Catalyst Additives to Enhance Mercury Oxidation and Capture

Final Report

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ABSTRACT

Catalysis is the key fundamental ingredient to convert elemental mercury in coal-fired power stations into its oxidized forms that are more easily captured by sorbents, ESPs, baghouses, and wet scrubbers, whether the catalyst be unburned carbon (UBC) in the ash or vanadium pentoxide in SCR catalysts. This project has investigated several different types of catalysts that enhance mercury oxidation in several different ways.

The stated objective of this project in the Statement of Objectives included testing *duct-injection catalysts, catalyst-sorbent hybrids, and coated low-pressure-drop screens*. Several different types of catalysts were considered for duct injection, including different forms of iron and carbon. Duct-injection catalysts would have to be inexpensive catalysts, as they would not be recycled. Iron and calcium had been shown to catalyze mercury oxidation in published bench-scale tests. However, as determined from results of an on-going EPRI/EPA project at Southern Research, while iron and calcium did catalyze mercury oxidation, the activity of these catalysts was orders of magnitude below that of carbon and had little impact in the short residence times available for duct-injected catalysts or catalyst-sorbent hybrids. In fact, the only catalyst found to be effective enough for duct injection was carbon, which is also used to capture mercury and remove it from the flue gas. It was discovered that carbon itself is an effective catalyst-sorbent hybrid.

Bench-scale carbon-catalyst tests were conducted, to obtain kinetic rates of mercury adsorption (a key step in the catalytic oxidation of mercury by carbon) for different forms of carbon. All carbon types investigated behaved in a similar manner with respect to mercury sorption, including the effect of temperature and chlorine concentration. Activated carbon was more effective at adsorbing mercury than carbon black and unburned carbon (UBC), because their internal surface area of activated carbon was greater.

Catalyst coating of low-pressure-drop screens was of particular interest as this project was being developed. However, it was discovered that URS was already heavily involved in the pursuit of this same technology, being funded by DOE, and reporting significant success. Hence, testing of SCR catalysts became a major focus of the project. Three different commercial SCR catalysts were examined for their ability to oxidize mercury in simulated flue-gas. Similar performance was observed from each of the three commercial catalysts, both in terms of mercury oxidation and SO_3 generation. Ammonia injection hindered mercury oxidation at low HCl concentrations (i.e., ~2 ppmv), yet had little impact on mercury oxidation at higher HCl concentrations. On the other hand, SO_2 oxidation was significantly reduced by the presence of ammonia at both low and high concentrations of HCl.

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INTRODUCTION

The objective of this project proposed by Southern Research Institute (Southern Research) was to investigate the enhancement of elemental mercury oxidation in coal-fired flue gas through catalysis. The first effort was focused on examining the catalytic nature of carbon types. Subsequently, an investigation of commercial SCR catalysts was conducted.

The behavior of SCR catalyst with respect to mercury is an extremely important issue for the utility industry at present. Evidence indicates that SCR catalysts have the capacity to oxidize mercury [1, 2]. Wet SO_2 scrubbers remove oxidized mercury efficiently, and utilities across the country are counting on future retrofits of SCR and wet scrubbers on their existing power plants to also allow them to meet any mercury-emission regulations they face. In fact, the Clean Air Mercury Rule (CAMR) was structured such that utilities could take advantage of the expected co-benefits of the SCR and FGD installations of the Clean Air Interstate Rule (CAIR).

Significant effort has been made to measure mercury oxidation across SCR reactors installed on full-scale power plants. However, the temperature, dust loading, and acid gases make mercury measurements challenging, especially for mercury-speciation measurement efforts. Specifically, dirty (ash laden) flue gas, high-carbon flyash, high acid-gas concentrations, saturated stack gas, and flue-gas temperatures as high as 800 °F (or as low as 250 °F) have made it very challenging to obtain accurate and validated mercury-oxidation and removal measurements. In fact, Ontario-Hydro measurements have been shown to be subject to significant sampling artifacts when made in these challenging environments, particularly from flue gas containing high-carbon ash and acid gases [3-5]. Nevertheless, some important information has been obtained from full-scale tests. Specifically, an EPRI Technical Report in 2002 concluded that residence time, catalyst temperature, and catalyst age all had a significant effect on mercury oxidation [6].

Some bench-scale work has been done, where, in the absence of ash, mercury measurements were significantly more effective, and the conditions, such as temperature and HCl concentration were much better controlled. These bench-scale studies have shown that mercury oxidation increases with increasing HCl concentration from 10 to 60 ppmv and that catalyst temperature has a significant effect on mercury oxidation between 550 to 770 °F [7, 8]. However, there is not much bench-scale data available, and the work performed has typically been limited to one type of catalyst, such as plate or honeycomb.

Modelers have had some success in predicting bench- and some full-scale data [9]. In fact, models have been able to squeeze a bit more information out of the limited experimental data that do exist from full- and bench-scale measurements. One of the models that have had success predicting mercury oxidation across SCR catalysts emphasizes a competing mechanism of HCl and NH_3 for catalyst surface sites [10, 11]. Both NO and Hg° were modeled as weak adsorbates on the catalyst surface, where high concentrations of NH_3 sustain NO reduction, yet inhibit mercury oxidation [10, 11].

The present SCR catalyst investigation compares three different types of commercial catalyst in a bench-scale apparatus, while examining the relationship of mercury oxidation to catalyst temperature, HCl concentration, and whether or not deNO_x is being implemented (i.e., with and

without ammonia). It is generally understood that typical vanadium catalysts for NO_x reduction tend to oxidize SO_2 to SO_3 in proportion to their ability to oxidize mercury, which can cause significant damage to a power plant and stack opacity problems. Hence, the three different catalysts were also compared for their SO_2 oxidation potential.

EXECUTIVE SUMMARY

The objective of this project proposed by Southern Research Institute (Southern) was to investigate the enhancement of elemental mercury oxidation in coal-fired flue gas through catalysis. The first effort was focused on examining the catalytic potential of different carbon types. Subsequently, an investigation of commercial SCR catalysts was conducted.

Originally, investigation of SCR catalysts was not intended to be part of this project. Rather, an investigation of stationary catalysts upstream of a wet scrubber was intended. However, during the development of the project, it was learned that stationary catalysts were well on their way to commercialization by URS. Hence, SCR catalyst investigations were added to the scope of work to supplement this low-pressure-drop screen work. As a result, the two main focus areas of this work were to (1) identify low-cost duct-injection catalysts to enhance mercury oxidation and (2) investigate the fundamental mechanisms governing mercury oxidation across SCR catalysts.

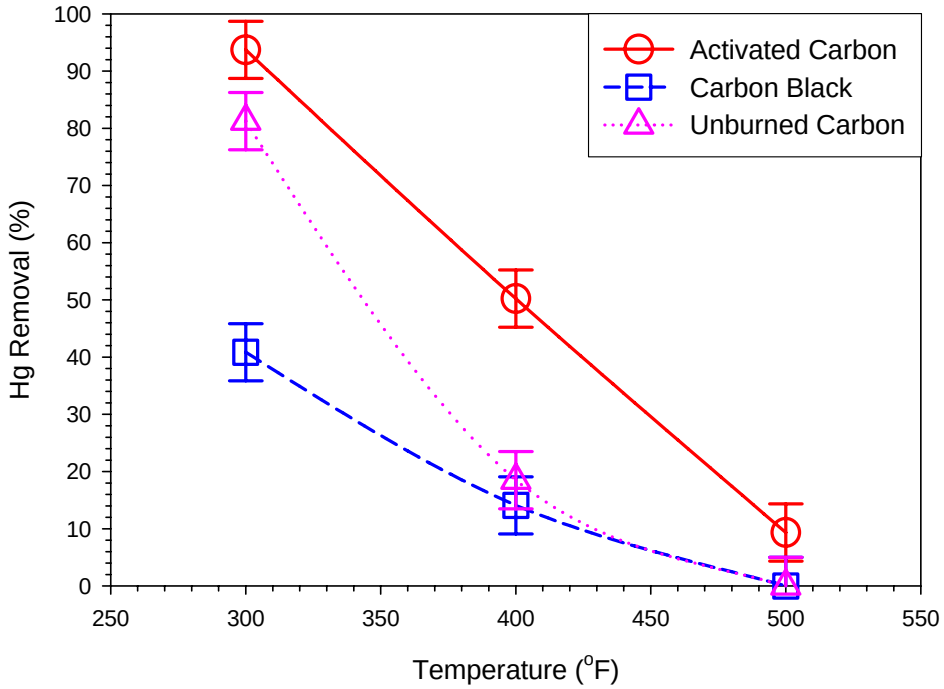
This project also benefited greatly from several existing fundamental investigations of mercury speciation and control that were conducted in the pilot-scale Combustion Research Facility (CRF) at Southern Research Institute. In fact, when this project was originally proposed, iron oxide and other cheap components found in flyash were considered as components to investigate in this work. However before final award, the existing projects determined that the only catalyst material inherent in coal flyash with a high enough activity to matter was unburned carbon. It had also been discovered that calcium had an enhancing effect on mercury removal, when carbon was present. Hence for this project, evaluations of duct-injection catalysts were concentrated on different carbon types and mixtures of carbon and calcium.

The Catalyst Test Facility (CTF) was used to derive fundamental kinetic information about each catalyst and catalyst material investigated for mercury oxidation and capture. At the core of the CTF is an extensive flue-gas simulation, gas flow, and metering system. The powdered carbon catalysts sat on a quartz frit that spanned the diameter of a 1.5” quartz reaction chamber, through which the entire gas flow passed. The SCR catalysts were held in place in the center of a 3” diameter reaction chamber, without obstruction. The clean (no particles) simulated flue gas contained all the major species present in real flue gas, including CO, CO_2, H_2O, O_2, N_2, HCl, NO, SO_2, SO_3, and Hg°, in concentrations that exist in the flue gases of existing power plants, burning specific coal types.

The simulated flue gas originated from compressed gas cylinders. The gases from the cylinders were then mixed to known concentrations by use of precision mass-flow controllers. The appropriate moisture content was generated through precise control of water evaporation. Mercury was added to the system with a PS Analytical 10.534 Mercury Calibration System, which consisted of a reservoir containing an inert substrate impregnated with elemental mercury maintained at constant temperature. The mercury reservoir supplied a saturated stream of

elemental mercury which was diluted before mixing with the other gases. The simulated flue-gas stream was well mixed and preheated before entering the reaction chamber. A 4 ½-inch diameter by 3-foot long tube furnace heated the reactor, which allowed the simulated flue gas to pass through the furnace while holding the catalyst samples in place.

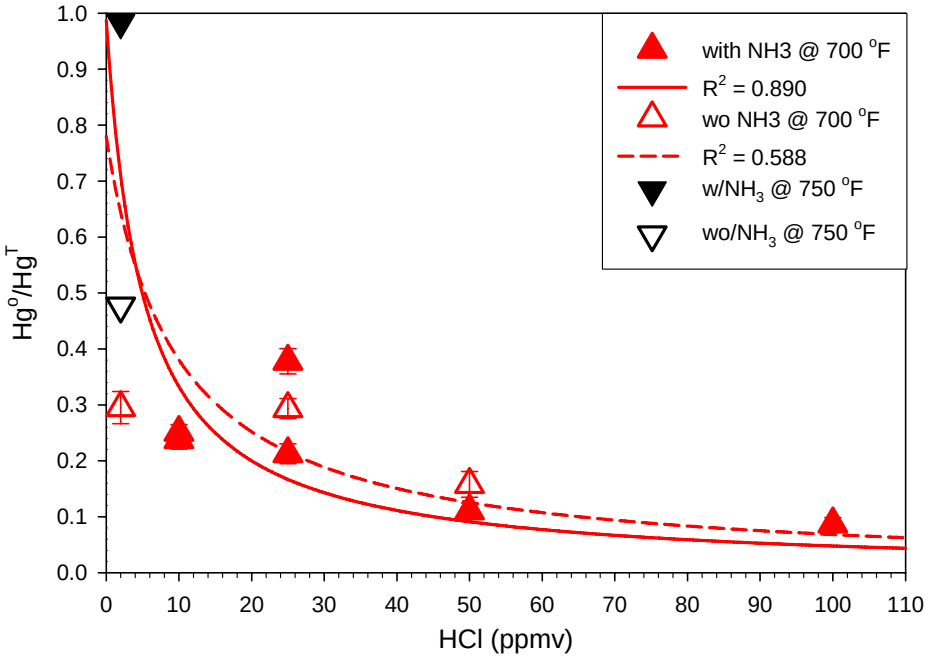
Bench-scale carbon-catalyst tests were conducted to obtain kinetic rates of mercury adsorption for different forms of carbon. The chemisorption rate of mercury on carbon was found to be first order in mercury concentration and half order in HCl concentration, for the facility configuration investigated. The applicable temperature range of the kinetic rates obtained is from 300 °F to 700 °F, and the applicable chlorine concentration range is from 2 ppmv HCl to 250 ppmv HCl. All carbon types investigated behaved in a similar manner with respect to mercury sorption, including their response to changes in temperature and chlorine concentration. As shown in the graph below for tests conducted with 50 ppmv HCl in the flue gas, activated carbon was more effective at sorbing mercury than carbon black and unburned carbon (UBC), because the internal surface area of activated carbon is greater than these other carbon types. The synergistic relationship between Ca and C is also discussed in the report.



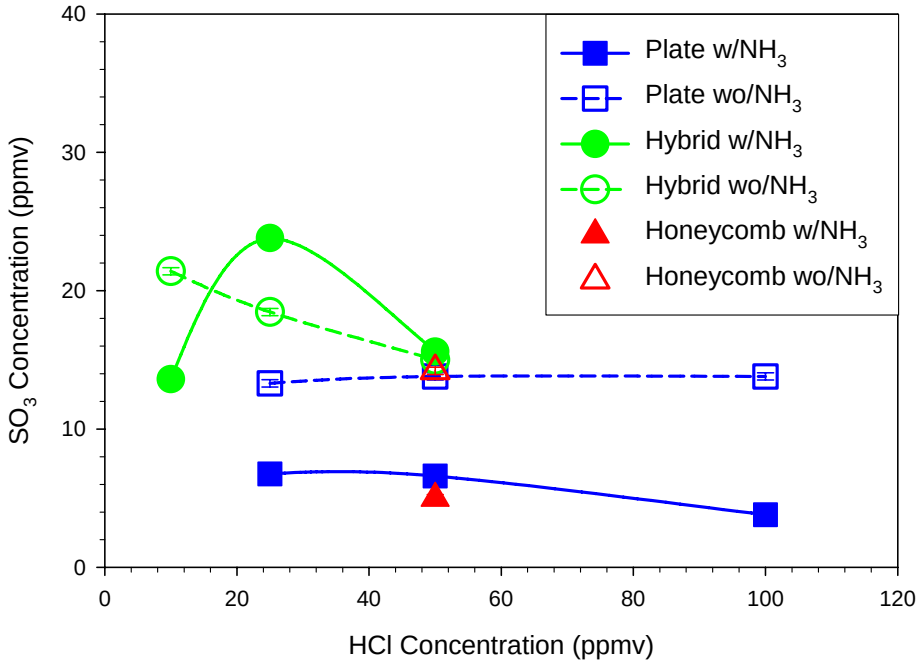
Selective Catalytic Reduction (SCR) for deNO_x has been shown to effectively oxidize mercury and thereby enhance mercury removal in wet scrubbers. Many years of research have gone into developing these catalysts for deNO_x, but relatively little was known about their mercury oxidation behavior. In this investigation, three different commercial SCR catalysts were examined for their ability to oxidize mercury in simulated flue-gas. The trade off between the advantage of mercury oxidation and the negative impact of SO_3 generation was also compared for the three catalysts.

The temperature range, area and space velocities, and bulk flue gas composition were consistent with conditions experienced at full-scale installations. The volume of each catalyst used was

chosen so as to provide a consistent $deNO_x$ value (with a one-to-one NH_3/NO ratio), for each catalyst. Quantitative information on the effect of HCl concentration, temperature, and ammonia injection on mercury and SO_2 oxidation are presented. Similar performance was observed from each of the three commercial catalysts, both in terms of mercury oxidation and SO_3 generation. The influence of $deNO_x$ (i.e., with or without ammonia injection) was mixed. Ammonia injection hindered mercury oxidation at low HCl concentrations (i.e., ~2 ppmv), yet had little impact on mercury oxidation at higher HCl concentrations, as shown in the figure below.



On the other hand, as shown in the figure below, SO_2 oxidation was significantly reduced by the presence of ammonia at both low and high concentrations of HCl.



Key conclusions obtained from this work are:

- When considering duct injection of catalysts or sorbents, laboratory-scale facilities, particularly fixed-bed experiments, operate at conditions that are much less representative of full-scale units than slipstream or pilot-scale units, such as the CRF, which possess a temperature-time profile that matches that of full-scale units.

- On the other hand, when considering testing of SCR catalysts, laboratory-scale facilities, such as the Catalyst Test Facility (CTF) used in this work, provide the most representative (and easy to control and measure) conditions of any test facility.

- The only duct-injection catalyst identified in this work was carbon, as a catalyst/sorbent hybrid.

- The most effective carbon type identified as an adsorbent or catalyst, alone or in a mixture with hydrated lime or limestone, was activated carbon, which is being widely used to remove mercury from existing coal-fired utilities.

- Elemental mercury is adsorbed onto SCR catalysts.

- SO_2 is adsorbed on to SCR catalysts.

- NH3 is adsorbed onto SCR catalysts and competes for vanadium pentoxide active sites with mercury and SO_2.

- HCl may be adsorbed onto SCR catalysts as well, but mercury, ammonia, and sulfur dioxide adsorption on SCR catalysts is much stronger.

- The presence of ammonia (i.e., in a normal 1-to-1 molar ratio of NO to NH_3) inhibits the oxidation of mercury across SCR catalysts, at low-HCl concentrations, consistent with low-chlorine PRB coals.

- At higher concentrations of HCl, ammonia has little effect on mercury oxidation.

- Ammonia was found to inhibit SO_2-to-SO_3 conversion across SCR catalysts, regardless of the HCl concentration.

- Concentration differences in HCl appeared to have little effect on SO_2-to-SO_3 conversion, at HCl concentrations between 10 ppmv and 100 ppmv.

- At low, PRB-levels of HCl, mercury oxidation increased with increasing temperature, from 650 °F to 800 °F. At higher concentrations of HCl, temperature differences within this range of temperatures had little effect on mercury oxidation.

- In addition to the information described in this list, because of the development that took place on this project in the ability to conduct mercury oxidation and SO_2-to-SO_3 conversion

tests across SCR catalysts, much more data and more precise and meaningful results were obtained during subsequent investigations in this area, particularly as part of a DOE University Coal Research (UCR) project. As a result of this subsequent work, combined with the data obtained in this work, a model describing mercury and SO_2 oxidation across SCR catalysts was developed, based on strongly adsorbed mercury, ammonia, and sulfur dioxide, and weakly adsorbed HCl, which model has been shown to effectively predict our data.

OVERVIEW OF OBJECTIVES AND TASKS

Objectives

The objective of this project was to identify different catalyst materials and means of using these catalysts to enhance mercury oxidation in coal-fired boilers systems. Specific interest in low-pressure-drop screens was initially in the forefront, when the project was proposed. However in the development of the final project, it was clear that low-pressure-drop screens or honeycomb catalysts were well on their way to the final stages of development, and it was also clear that there was a significant need to investigate SCR Catalysts for mercury oxidation and SO_2-to-SO_3 conversion. Hence, the investigation of different commercial SCR catalysts became a major objective of the project to supplement the investigation of low-pressure-drop screens.

Furthermore, it was determined that the mercury-oxidation data could best be obtained in the Catalyst Test Facility (CTF), meaning that the CTF was the best facility at reproducing the conditions that SCR catalysts would experience at full-scale coal-fired power plants. Hence, the objective of testing catalysts in the pilot-scale Combustion Research Facility (CRF) became much less of a priority. Testing in the CRF was reserved for particular catalyst-implementation strategies, which turned out to be primarily, injection of carbon as a catalyst/sorbent hybrid. In addition, a major objective of this project was to identify and describe mechanisms responsible for the catalytic mercury oxidation, experimentally observed. Data from this work was provided to modelers independently developing mercury-oxidation models across SCR catalysts, and subsequently improvements to these fundamental models were made, which are now just being published. In addition, global rate models were developed to describe and predict mercury adsorption rates on chlorinated carbon sites (the first step in carbon-catalytic oxidation of mercury), and these models are contained in this report.

Task 1: Development of Experimental Matrix

Task 1.1: Identification of Potential Catalysts

For this project, Southern Research's existing knowledge base, information obtained from the on-going EPA/EPRI Chemistry of Mercury Speciation (CMS) Project and a concurrent DOE Ca-Based Sorbents (CBS) Project ("Mercury Control with Calcium-Based Sorbents and Oxidizing Agents", DE-PS26-01NT41183), along with information in the literature was used to identify catalysts and implementation strategies. The EPA/EPRI CMS Project was an on-going project lasting approximately 5 years, under which Southern Research investigated the fundamental

mechanisms (other than those related to SCRs) governing mercury speciation in coal-fired power systems. The DOE CBS project also involved fundamental research at the pilot scale, where alternatives to activated carbon (particularly Ca-based sorbents, but also reagents such as Na_2S_4 and different forms of halides) were evaluated for control of mercury. Among other things, the extensive efforts of the DOE CBS project led to an understanding of the role of calcium (either from flyash or introduced through sorbents) on mercury speciation and adsorption in coal-fired power systems.

When the "*Catalyst Additives to Enhance Mercury Oxidation and Capture*" project was proposed, iron, calcium, and other metals in flyash were considered possibilities for potential catalysts. By the time this project was awarded, information from the on-going tests conducted in the CRF showed that iron and calcium were not effective mercury-oxidation catalysts in the flue-gas environments of coal-fired utilities. In fact, it was found that the only catalyst inherent in coal ash that had a very significant effect on mercury oxidation was unburned carbon (UBC). Hence, for duct-injection catalysts, a number of different carbon types were identified, such as carbon black, activated carbon, and unburned carbon.

As mentioned earlier, it became clear that the concept of low-pressure-drop screens or honeycomb catalysts upstream of a wet scrubber had already been significantly developed by DOE and URS. Additionally, URS had already tested gold and palladium as catalysts and developed forms of these catalysts that were effective for the honeycomb application. While they had spent significant efforts with carbon-based catalysts for a lower-cost option, they were not able to identify a carbon catalyst that worked well in the flue-gas environments in which it would need to work. As a result of the efforts and findings described above, it was determined that there were not any catalysts for Southern Research to test on this project for use in low-pressure-drop screens. Furthermore, it was determined that gold, palladium, iron, and calcium catalysts were not favorable for testing or use as duct-injection catalysts. Only carbon, in different forms, was identified as a material suited for duct injection, and due to the nature of carbon it was clear that carbon was a catalyst-sorbent hybrid, i.e., activated carbon was already used as a mercury sorbent.

The investigation of SCR catalysts became the most significant objective of this project, as a result of the project events and analysis described above. Many years and dollars have gone into the development of SCR catalysts, so it was reasonable to choose from existing commercial SCR catalysts. Three commercial SCR catalysts were chosen for this work, a honeycomb-type Cormetech catalyst, a plate-type Hitachi catalyst, and a corrugated-type Haldor Topsoe catalyst, which is in appearance a hybrid of a plate and honeycomb catalyst. Samples of each type of catalyst were donated to Southern Research for this project. However, they agreed to do so on the condition that the specific details of their catalyst and manufacturing process would not be revealed. Hence, we cannot provide all the specifications on these catalysts. However, each catalyst was designed for deNOx, based on the same application criteria, and each was a standard formula, not specifically designed for low SO_2-to-SO_3 conversion. We also know that each catalyst support contained titanium and that the active material in each catalyst is vanadium pentoxide, which is the same active catalyst for mercury and SO_2 oxidation.

Task 1.2: Bench-Scale Catalyst Optimization

The bench-scale testing took on a much bigger role in this project than originally anticipated. The catalytic properties of different forms of carbon and calcium were indeed investigated in the bench-scale unit for potential subsequent testing in the CRF. However, with the addition of the SCR catalyst testing and the decision that the bench-scale facility produced the most realistic testing conditions for these tests, most of the testing took place in the bench-scale Catalyst Test Facility (CTF). In fact, over several years, the primary focus of the CTF was to obtain the data necessary to fulfill the objectives of this project. In addition to testing the three catalysts mentioned, flue-gas components relative to different coal types were also examined. Specifically, the impact of hydrochloric-acid concentration on mercury and SO_2 oxidation was examined, over a range of differences in interacting parameters, such as temperature, residence time, NO concentration, and the NH_3/NO ratio.

The information obtained from the SCR catalyst evaluations has been published and provided to external modelers for use in developing and validating their models. Fundamental insights into the governing mechanisms were obtained. In addition, the data from this work and the equipment and trained personnel established in this work has led to follow-on work in this same facility that has furthered the understanding of mercury and SO_2 oxidation across SCR catalysts. In fact, a new model has recently been published by Southern Research and the University of Alabama at Birmingham (UAB) research scientists, based on the data obtained in this work and subsequent testing funding by the UCR project. This new model takes some of the approach used by existing modelers and improves upon these mechanisms, based on the added understanding obtained in this and the UCR work.

Task 1.3: CRF Experimental Design

As mentioned above, when the proposal was first submitted, it was envisioned that this project would proceed toward the demonstration of a low-pressure-drop screen covered with catalyst material just upstream of a wet scrubber. However, by the time this project was awarded, results from other projects warranted that the focus of this project be changed to investigating SCR catalysts and the catalytic properties of carbon, relative to mercury. This shift in the focus and subsequent confirmation of this approach coupled with the need to perform SCR catalyst testing in the CTF, resulted in most of the testing being done in the bench-scale facility. However, CRF tests were conducted that provided valuable information to this project, including the fact that carbon was by far the most significant catalyst inherent in coal flyash (iron and calcium having only a small insignificant impact). All the information that enabled the elucidation of the mechanism of mercury adsorption and oxidation on carbon and the role that calcium plays in the enhancement of mercury capture by carbon was also obtained through CRF testing. These experiments in the CRF however, were conducted as part of the EPA/EPRI CMS project, aided by data obtained on this project in the CTF. Therefore, the need to develop a CRF experimental design was eliminated.

Task 2: Mercury Speciation Investigation

Task 2.1: Learn from Larger Mercury-Speciation Investigation

Conducting the EPA/EPRI CMS pilot-scale project prior to and during this project was indeed a great help to this project. As already mentioned, an understanding of mercury oxidation in coal-fired boilers (absent of SCRs) was elucidated in the EPA/EPRI CMS investigation, and the relative importance of iron, calcium, unburned carbon (UBC), and other ash minerals was determined in that work. Hence, unlike conventional research, as far as the non-SCR mechanisms are concerned, the fundamental research took place in the pilot-scale facility as a precursor to later bench-scale work to investigate specific question, such as the adsorption rate of mercury on different carbon types. This was appropriate, because of the complicated nature of mercury speciation.

Bench-scale, packed-bed, long-time experiments on flyash, carbon, and sorbents would not be at all representative of the conditions that particles and mercury experience in the approximately 7 seconds from the time pulverized coal is injected into the burner of a full-scale power plant and the exhaust leaves the stack. Because of the volatility of mercury and its low concentration, it tends to be greatly affected by small changes in temperature, time, and gas composition. Therefore, it was important to conduct this fundamental mercury speciation investigation at the pilot-scale, where conditions could be realistic and where parameters could still be carefully controlled.

The CRF does not have the capability of testing with SCR catalysts, and as it turns out, the kinetics of reactions involving SCR catalysts are best investigated at bench-scale, because SCR catalyst take a long time (up to two full days), to obtain steady-state absorption with respect to mercury and SO_3. In addition, SCR catalysts in full-scale boiler units are stationary in the duct as flue gas passes through them, just as they were in the bench-scale CTF.

Task 2.2: Learn from Larger Investigation on Sulfuric-Acid Emissions

Although it was indeed a goal of the EPA/EPRI CMS project to investigate SO_3 formation and control, most of the project was focused on mercury speciation, primarily because mercury was more difficult to measure, had a more difficult chemistry, and was more difficult to understand. The impact of SO_2 on mercury oxidation (absent an SCR) was investigated in the pilot-project. While EPRI and EPA have since funded modeling work at Southern Research, building on a long history of both experimental and modeling work on SO_3 that has helped to answer remaining questions about SO_3 formation and control, little information was available from this particular project on this subject, at that time. Instead, SO_3 generators were used in the CTF to examine the impact of SO_3 on carbon deactivation, which was the only mechanism observed whereby SO_3 affected mercury oxidation. The impact of SO_2 on mercury oxidation across SCR catalysts was also investigated in the CTF.

Task 2.3: Contribute to the Larger Mercury-Speciation Investigation

This project contributed to the EPA/EPRI CMS project by directly comparing the adsorption rate of different carbon types, which was easier and cheaper in the bench-scale facility, and by investigating mercury speciation across SCR catalysts. Slipstream test facilities for SCR catalysts are difficult facilities in which to investigate fundamental mercury chemistry, because flue gas composition, flow, and temperature changes so much at actual power plants. Given that it may take a day or two for a catalyst to fully come to steady state after a significant temperature or acid-gas composition change, fundamental examination of mercury across SCR catalysts is much more suited for bench-scale or micro-scale units.

Task 2.4: Parametric Testing

Extensive parametric testing took place in the CTF, first on carbon adsorption and subsequently on SCR catalysts, relative to mercury and SO_2 oxidation as a function of catalyst type, HCl concentration, temperature, residence time, and ammonia and NO concentration. Subsequent testing in the CTF on the effect of CO on mercury oxidation has now also been published, which was funded by the DOE UCR project. In all of the parametric testing, the first step was to establish an effective and appropriate method for performing the tests and measuring the necessary components at the inlet and outlet of the reactor, such as HCl, $HgCl_2$, Hg, and SO_3. This was done, and the subsequent parametric testing was performed.

Task 3: Modeling

Task 3.1: Fundamental Mercury-Speciation, Rate Models

As part of the DOE CBS project, which also partnered with the EPA/EPRI CMS project, a fundamental mercury-speciation, rate model was developed that predicts changes in mercury speciation from the coal in the burner to the particulate collection devices, in the absence of an SCR. The model to date is effective, but it could use additional data on the relationship between carbon-type activity toward mercury as a function of carbon burnout and coal type, in order to more exactly predict mercury speciation and capture for any power station. Each and every external model also needs this added input, if more accurate predictions are to be made. The testing of mercury adsorption on different carbon types added to the development of this model, which was published in a Masters Thesis [28] and also in a MEGA Symposium [15].

All of the data obtained in this work was provided to the external modelers as described in the SOPO, which they used to help develop and validated their models. NEA chose to develop their SCR model entirely based on data from full-scale plants, however. Furthermore, while REI produced a fundamental model for SCRs based on a different approach, it also was not able to include all of the observations made in this work. Consequently, another model has recently been developed by UAB and Southern Research, based on the data obtained in this work and additional data obtained in the CTF, which uses mechanisms similar to that of NEA and REI, but has improved upon the mechanism. The result is a model that not only predicts the data from this work, but more effectively predicts mercury speciation across full-scale SCR units. This model has been recently published elsewhere [12, 13] and was not part of this work.

Task 3.2: Semi-Empirical, Rate Model

An objective of the project was that semi-empirical rate models may be developed to describe the parametric relationship between mercury oxidation and the addition of catalyst materials. As the project progressed, certain implementation options were eliminated and others were elucidated. In the end, most of the modeling performed (albeit outside of funding for this project) was fundamental. Fundamental models were developed containing the mechanisms necessary to describe mercury speciation in coal-fired power stations, other than the impact of SCRs, and separately, fundamental models of mercury oxidation across SCRs were developed. Where fundamental models were developed that could accurately predict mercury speciation, semi-empirical models were not needed. However, one such global rate model was developed to allow an effective and simple comparison of the rates of mercury adsorption by different carbon types. This model is presented in this report (see Eqs. 3 and 4), along with rate constants for each of the carbon types (see Table 2).

Task 4: Design of Applicable Mercury-Oxidation Catalysis Processes and Equipment

The level of effort planned for this task consisted of the design of equipment or processes for specific applications based on the information obtained in the first three tasks. This task was not intended to involve any construction or testing. Based on the information obtained from these three tasks, the design of equipment was not necessary. Injection systems for activated carbon are already in the market place, and SCRs have been commercial for years. Low-pressure-drop honeycomb catalyst units upstream of wet scrubbers have also been built and tested at full-scale power plants.

Task 5: Reporting and Technology Transfer

Quarterly Technical and Financial Reports were required on this project, in addition to this Final Technical Report and the financial and other final reports required. All of the required reports have been produced and delivered to DOE in accordance with the reporting requirements. In addition, several presentations and technical papers have been presented, and these references have been listed in the bibliography. Also, each paper presented at a technical conference or otherwise published was sent to the DOE COR for this project. The models, mechanisms, and data obtained from this project have all been published in DOE reports and relevant technical conferences.

EXPERIMENTAL

Figures 1 and 2 show pictures of the CTF's quartz furnace (micro-reactor), gas-conditioning bubblers for mercury speciation and stabilization prior to mercury monitoring, flue-gas continuous emission monitors (CEMs), and gas-flow control systems. Both elemental and total mercury were measured at the outlet of the CTF. The CTF simulated clean (no particles) flue gas with all the major flue-gas species present, including CO, CO_2, H_2O, O_2, N_2, HCl, NO, SO_2, SO_3, and Hg°, in concentrations that exist in the flue gases of existing power plants, burning specific coal types.

Hot Reactor Sorbent in Reactor SCR Catalyst in Reactor

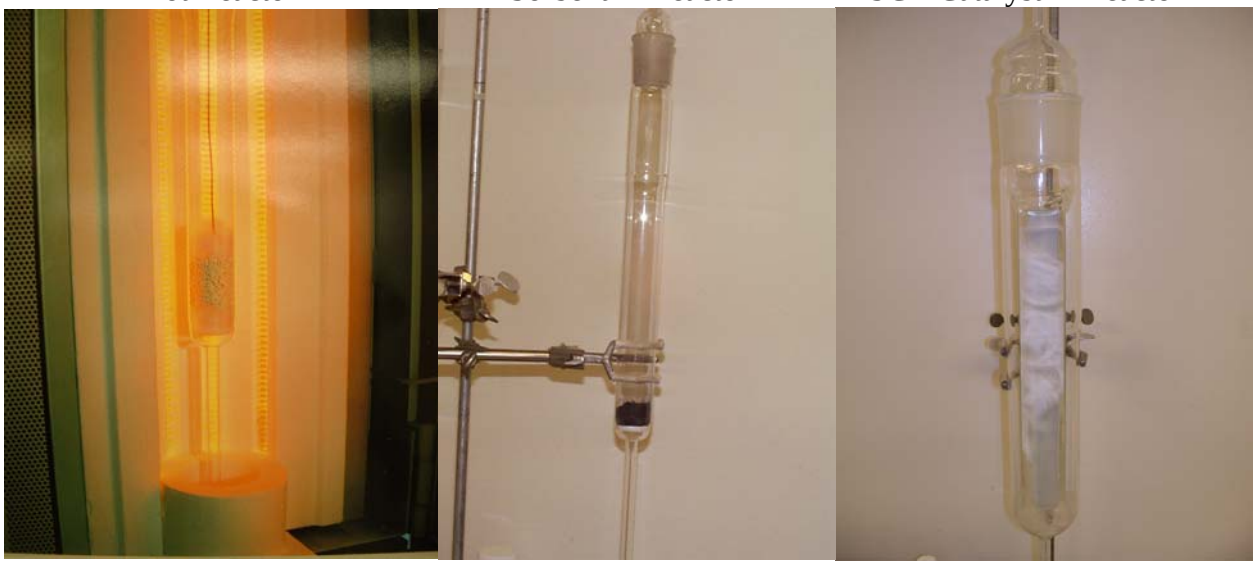


Figure 1. CTF quartz furnace at ~1000 °C (left) and outside of furnace (middle and right).

Figure 2. CTF furnace, gas-injection system, flue-gas CEMs, and mercury gas-conditioning system.

The simulated flue gas originated from compressed-gas cylinders. The gases from the cylinders were then mixed to known concentrations by use of mass flow controllers. Evaporating liquid water generated the appropriate moisture content in the gas stream, and mercury was added to

the system as elemental mercury vapor carried in a clean air stream. The simulated flue-gas stream was well mixed and preheated before entering the reaction chamber. A 4 ½-inch diameter tube furnace heated the 3-ft long tubular reaction chamber, which carried the gases through the furnace while holding the catalyst samples.

All heated sections of the micro-reactor within the CTF system were made of quartz glass or Pyrex to limit wall effects. A semi-continuous emission monitor (SCEM) was employed to detect the mercury levels exiting the reaction chamber. A gas-conditioning system was used to convert all mercury into the elemental form, for detection using a combined gold-trap and atomic fluorescence monitor. A Tekran Model 2573A Mercury Vapor Analyzer was used to detect the elemental mercury. Along with mercury, simultaneous measurements of oxygen, carbon dioxide, nitrogen oxides, and sulfur dioxide were made using continuous emission monitors.

Figure 3 shows a schematic of the CTF system layout. The flow rates, temperatures, and concentrations were continuously monitored and maintained throughout the test program.

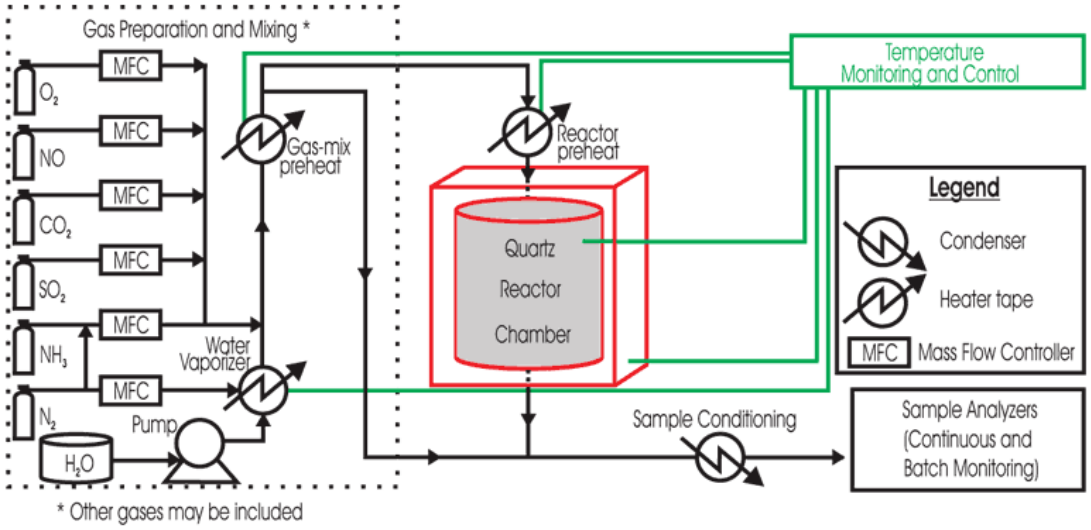


Figure 3. CTF gas-flow system.

Carbon Catalysts

The carbon catalyst tests in the CTF were designed to simulate conditions relevant to mercury adsorption onto chlorinated carbon sites on carbon particles disperse in coal-fired flue gas. The adsorption step is the first step in the oxidation of mercury in coal-derived flue gas. Hence, it was desired to isolate this step and obtain the rate of adsorption of mercury on unburned carbon (UBC) and other carbon types. In doing so, the experiments were designed to yield primarily adsorption observations, without the confounding additional observation of mercury oxidation, which would have made the data much more uncertain, less repeatable, and difficult to interpret.

Breakthrough tests have been conducted elsewhere that allow adsorption of mercury onto the carbon bed until the carbon is saturated with mercury and subsequently begins to release mercury

(oxidized or not). Breakthrough tests are not representative of the mechanism whereby mercury adsorbs onto carbon in flue gas and subsequently oxidizes, because dispersed carbon particles in flue gas are not saturated with mercury. In fact, breakthrough tests on baghouse filter cakes are also not representative of adsorption of desorption of mercury on carbon, because the carbon on filter cakes (whether the carbon comes from the flyash or activated-carbon injection) is continually renewed by incoming carbon and discharged when the baghouse is pulsed. Therefore, the tests for this work were designed to examine mercury adsorption on carbon with significant available carbon surface area, which is consistent with full-scale conditions.

Carbon catalyst samples were put into the quartz reactor with a quartz frit and quartz filter paper to prevent the sample from contaminating the gas-flow system. The bed depth of the catalysts was approximately one-third of an inch in the direction of gas flow. The precise bed depth was measured for each test. At the beginning of each experiment, a blank quartz reactor was inserted into the gas stream to collect baseline data. After sufficient data were collected, the blank reactor was removed and immediately replaced with the catalyst-packed reactor. The reactor was allowed to come to the initial 149 °C (300 °F) temperature, at which time the exposure experiment began. Subsequently, higher temperatures were investigated, up to 594 °C (1100 °F). At the conclusion of each experiment, the quartz filter paper and exposed sample was disposed of and the quartz reaction chamber was thoroughly cleaned and repacked with the next powder to be tested. These steps were precisely repeated for each material tested.

SCR Catalysts

Three types of SCR catalyst were examined in this work, plate, honeycomb, and hybrid. Hitachi Corp. provided the plate catalyst. Cormetech Inc. provided the honeycomb catalyst, and Haldor Topsoe Inc. provided the hybrid catalyst, which is a hybrid of the plate and honeycomb geometries. Each type of catalyst was derived from a distinctly different manufacturing process. Examples of the three different catalyst types are shown in Figure 4. Each of these three catalyst types were designed for the same commercial application of SCR for deNO_x. The majority (i.e., over 95%) of NO_x in the flue gas of coal-fired boiler systems is in the form of NO. Hence, NO was exclusively used in this work to simulate the NO_x concentrations in the flue gas.

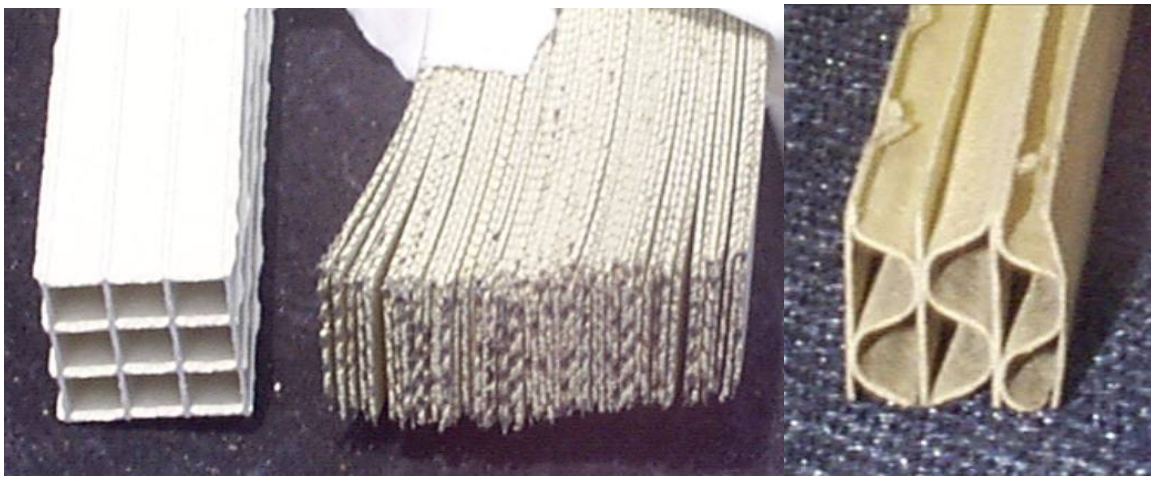


Figure 4. From left to right, honeycomb, plate, and hybrid SCR catalysts.

While these three catalysts are commercial catalysts, all of the details concerning the structure of each catalyst and the manufacturing process are not published. However, it is known that each catalyst substrate contains titanium and that the active component of each catalyst is vanadium pentoxide.

RESULTS AND DISCUSSION

Carbon Catalyst Investigation

Carbon Catalyst Test Parameters

Four types of carbon were examined in this test matrix, unburned carbon (UBC), FGD activated carbon (AC), carbon black (CB), and soot. The UBC was obtained from separating the residual carbon from bituminous-coal flyash. This process resulted in enriching the UBC to over 50%. However, a significant amount of ash remained with the UBC, thus diluting the carbon content, which should lead to less mercury sorption than for the activated carbon.
Bituminous-coal flyash is generally higher in carbon than PRB flyash, and the UBC is primarily contained in separate carbon particles, whereas PRB-flyash UBC is generally intimately associated with its flyash mineral matter. Coal-blending and ash-injection investigations previously conducted at Southern Research have shown that UBC in ash has a significant effect on both mercury oxidation and capture [3, 4, 14, 15], which is enhanced by the calcium present in PRB flyash. In fact, it was found in the previous work that the removal of both elemental and oxidized mercury by UBC was enhanced by the presence of calcium in PRB flyash and in calcium-based sorbents [3, 4, 14, and 15]. Hence, enhancement of mercury-oxidation and capture by different forms of carbon with several different forms of calcium were also examined in this test matrix.

The CTF at Southern Research was used to conduct all bench-scale experiments in this work. The quartz reactor was filled with either 1.5 or 3 grams of sample in a packed bed for each test, depending on the expected rate of sorption. Ultimately, the samples were compared using a common gas-sorbent contact time, as described in the next section. This contact time was based on the average bulk residence time of gas passing through the sorbent bed, rather than on a total surface area basis (considering pore diffusion times), because surface area was considered an inherent property and difference of each carbon type, and because practical application of these carbons will be compared on a cost/mass basis.

Normalizing to Comparable Residence Time

Each carbon type or calcium/carbon mixture tested had a different gas-contact time, which was a function of the density and compactness of material and sample. For example, activated carbon was generally very light, and thus did not pack down. This allowed a significant void volume within the carbon bed through which the simulated flue gas could flow. On the other hand, limestone, which is very dense, packed down tight, leaving much less void volume for the gas to flow through. This difference in void volume directly affects the comparability of samples, because the gas-carbon contact time is proportional to the void volume within the material bed. In order to place the results of each carbon and carbon/calcium material on a comparable basis,

the mercury removal values were adjusted to a common gas-carbon contact time (residence time), using the first order rate law.

Assuming that the reaction of mercury on the carbon sites is first order with respect to mercury concentration, and that the concentration of carbon sites is an inherent property of the system that does not change with time, then the concentration change of mercury across the reactor bed can be described by:

$$\frac{dC_{Hg}}{dt} = -kC_{Hg} \qquad (1)$$

where the initial and boundary conditions are respectively:

$@\ t = 0.0 \qquad C_{Hg} = C_{Hg}^{o} \qquad$ and $\qquad @\ t = \infty \qquad C_{Hg} = 0.0$

Hence, the fraction of mercury removed from the simulated flue gas as a function of time is:

$$X_{Rem} = 1\text{-}X_{Hg} = 1\text{-}e^{-\alpha t} \qquad (2)$$

where α is a constant, independent of time and the fraction of mercury removed. A value of α was determined for each test condition, based on the residence time and fraction of mercury removed. Then, using that calculated-α value, the fraction of mercury removed was normalized for all conditions to a consistent residence time of 0.12 seconds, which was a calculated average of gas-carbon contact times for all experiments conducted in this specific investigation. This was a reasonable approach, because the total range of residence times measured was between 0.08 and 0.21 seconds.

The residence times were determined for each condition by dividing the bed-void volume by the gas flow rate through the bed. The void volume was obtained for each condition by subtracting the total particle volume from the total volume taken up by the carbon (or other powder) in the reactor bed, using the following particle densities for each material (see Table 1).

Table 1. Particle densities and surface areas for carbons and limes tested in CRF.

Powder Type	[a]Particle Density (g/cc)	BET Surface Area (m^2/g)
Activated Carbon	0.77 +/- 0.22	600
Hydrated Lime	~2.2	18
Limestone	2.2 – 2.8	--- [c]
Carbon Black	~0.75	120
Unburned Carbon	~0.76	20
Soot	---[b]	62

a. Values provided by manufacturer or supplier.
b. Soot particle densities can range from 0.3 to 1.8 g/cc [16, 17].
c. Typical values range from 2 to 5 m^2/g, for ground limestone (80% passing 100 mesh) [18].

Relevance to Full-Scale Residence Times

The flue-gas contact time with the baghouse dust cake in full-scale baghouses ranges from approximately 0.09 seconds to 0.33 seconds, for the range of baghouse technologies and applications that exist in the United States, from reverse-gas and shaker-baghouses to COHPAC pulse-jet baghouses. Thus, the 0.12 seconds of gas/sorbent contact time in the CRF is within the range of full-scale baghouses, albeit on the low end of the range. Furthermore, full-scale dust cakes are a mixture of carbon and flyash when activated carbon is injected. The inclusion of fly ash adds mostly inert material (i.e. silica, alumina, and iron) reducing the total concentration of carbon in the dust cake, thus effectively reducing the flue-gas contact time with the active material. Hence, the low end of the residence time range is the most appropriate to use for relevance to full-scale conditions.

Carbon Catalyst Kinetics

Initial tests compared the capture of mercury by four different carbon types, activated carbon, carbon black, unburned carbon (UBC), and acetylene soot (used to represent the nature of soot in general). From a qualitative comparison, it was determined that UBC and activated carbon were similar in reactivity and yielded more mercury capture than carbon black or soot. Soot has few nodes or active carbon sites where reactions can take place. In general, soot is difficult to oxidize once formed. These qualitative results indicated that soot is also less reactive with mercury. From the qualitative results, it was clear that soot was very unreactive and would not effectively adsorb mercury. Therefore, soot was not pursued in the quantitative analysis.

The quantitative comparisons are shown in Figures 5-8, with +/- 5 $\mu g/m^3$ error bars, representing the general uncertainty of the data and mercury measurements, which was established in earlier repeat tests in the CTF with the mercury monitor. The data from tests conducted that clearly had greater error, due to a leaking or contaminated system, poor temperature control, or problems with instrumentation, were discarded. More descriptive error bars would require multiple repeats of each test condition with subsequent statistical analysis, such as establishing confidence intervals. However, the data and given uncertainties were sufficient to make the assessments made in this report. Given the nature of the results obtained and the conclusions drawn, it would not have been the best use of funds and time on this project to obtain multiple repeats, since the conclusions did not lead to a potential breakthrough in catalysts for mercury removal.

Figures 5-8 illustrate that mercury capture was much more effective at 300 °F than at higher temperatures. In addition, mercury removal was more effective with higher HCl concentrations. While the activated carbon was most effective at removing mercury, the UBC, with a significant but smaller internal surface area (see Table 1), was also effective at removing mercury. A curve for soot would lie on the bottom of each of the figures.

The data for this section of the report may be found in Tables A.1 through A.3 of the Appendix.

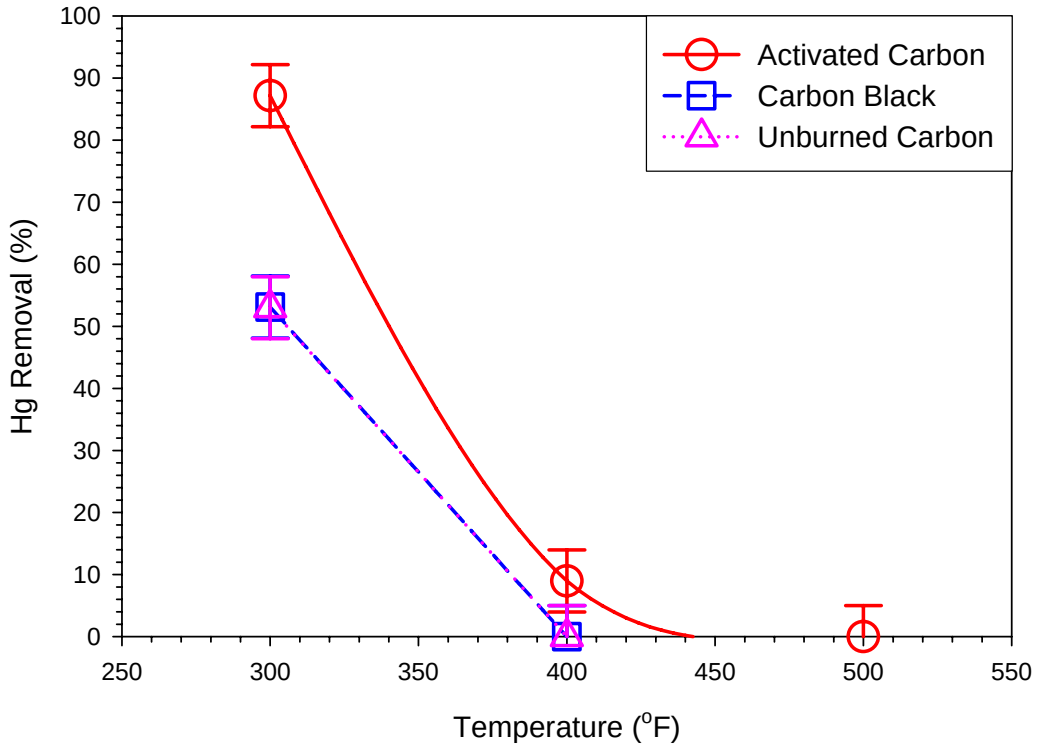


Figure 5. Three C-types compared in simulated flue gas with 2 ppmv HCl.

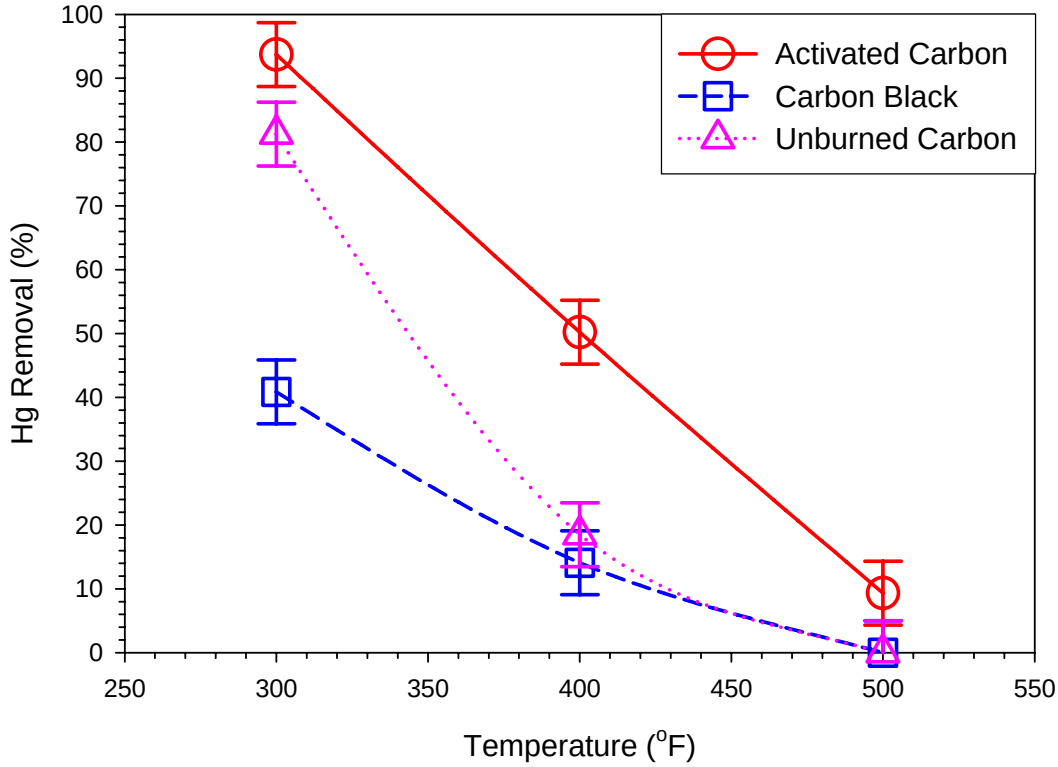


Figure 6. Three C-types compared in simulated flue gas with 50 ppmv HCl.

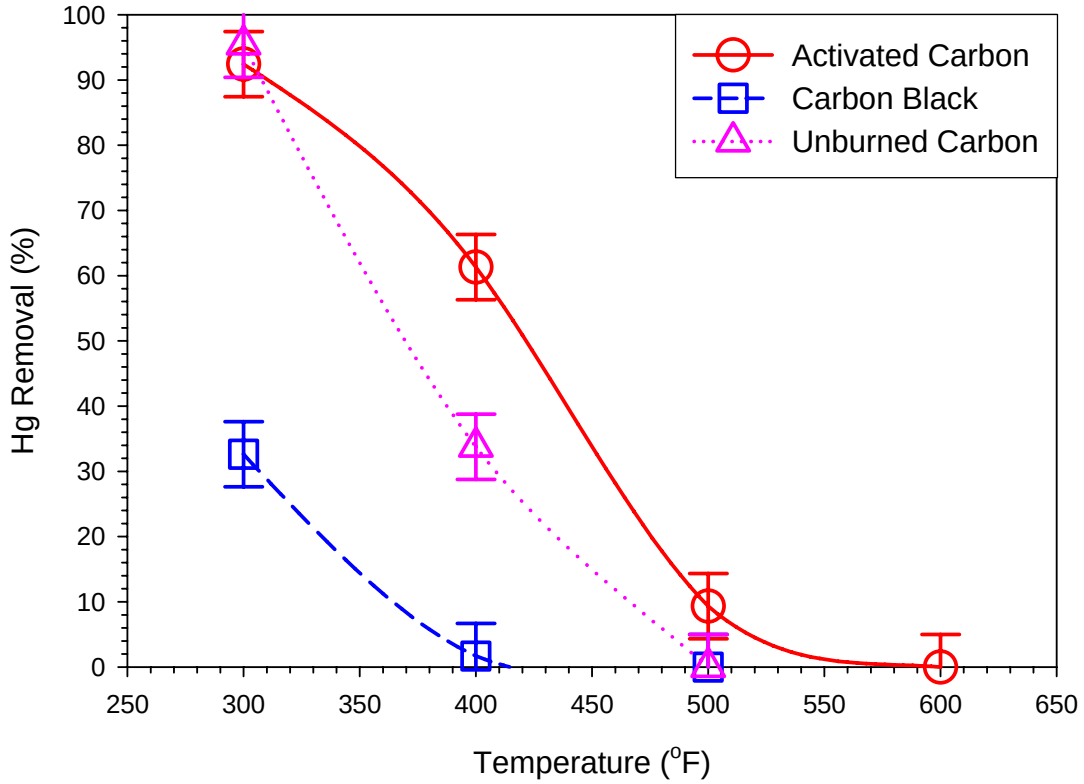


Figure 7. Three C-types compared in simulated flue gas with 100 ppmv HCl.

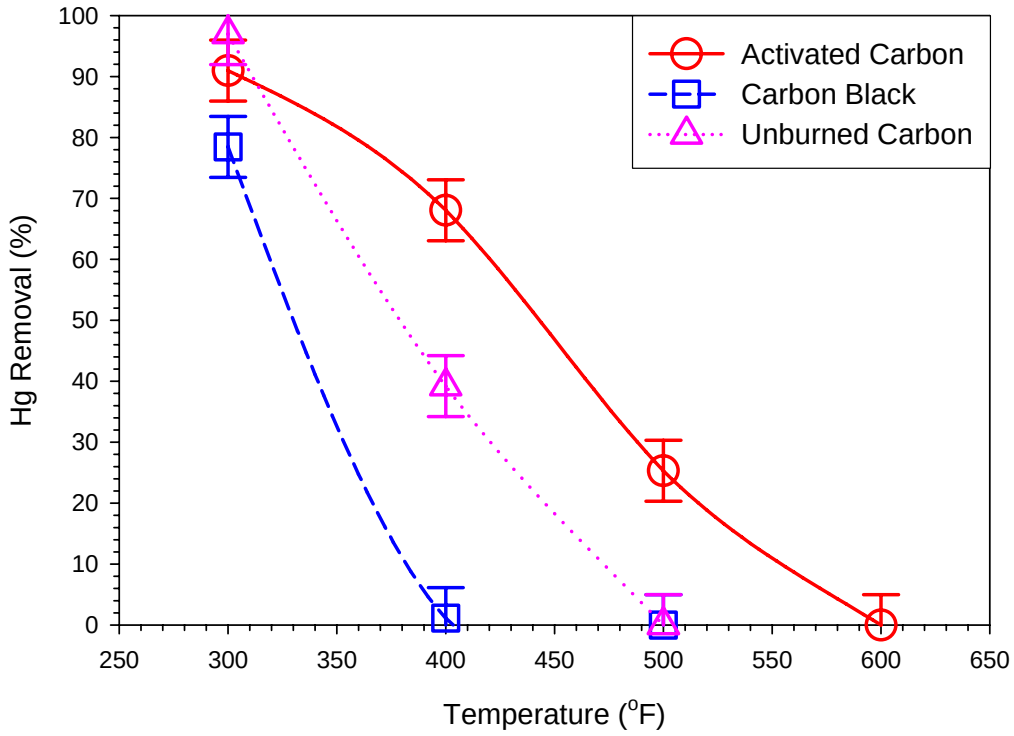


Figure 8. Three C-types compared in simulated flue gas with 250 ppmv HCl.

The separation between measured total mercury and measured elemental mercury was small and changed little, regardless of the flue-gas condition or bed material. Hence, it was not possible to directly isolate and consider the oxidized mercury separate from the removal data. However, the mechanisms by which mercury is oxidized and removed from flue gas are related.

The first step in mercury oxidation in the duct of a coal-fired-boiler back pass is for HCl to adsorb onto UBC in the ash particles, forming chlorinated carbon sites [19-21]. Mercury then reacts with the chlorinated carbon sites to form HgCl on the surface of the carbon sites. Next, the oxidized mercury may desorb back into the flue gas, where it may oxidize further to $HgCl_2$. Finally, $HgCl_2$ may readsorb onto open carbon sites. The concentration and availability of open carbon sites determines the rate of mercury adsorption and desorption. If there is an abundance of carbon, then mercury adsorption will dominate over desorption, resulting in a net mercury removal. If carbon sites are scarce, desorption may be nearly as rapid as adsorption, in which case very little mercury removal will occur, but significant mercury oxidation may take place [3, 15, 19-21]. This mechanism was quantified and described as part of the EPRI/EPA CMS project and DOE CBS project discussed earlier [3, 22]. The details of this mechanism have recently been published in Fuel Processing Technology [23].

Since the mercury oxidation pathway is intimately linked with the mercury adsorption process, the removal data taken in the CTF can be used to develop quantitative information regarding mercury oxidation, as well as mercury capture. To this end, quantitative reaction constants for mercury adsorption were derived for each of the three major carbon types investigated in the CTF – activated carbon, carbon black, and unburned carbon (UBC). The reaction of mercury with chlorinated carbon sites is first order with respect to mercury concentration. The order of the reaction in terms of HCl in the flue gas was unknown, since this depends on the process of forming chlorinated carbon sites, prior to reaction with mercury. Hence, the following equation was used to describe the reaction rate:

$$\frac{dC_{Hg}}{dt} = -k_{Hg,HCl} C_{Hg} C_{HCl}^{n} \qquad (3)$$

where C_{Hg} and C_{HCl} represent the concentration (mol/cc) of elemental mercury and hydrochloric-acid vapor in the simulated flue gas. The order of the reaction in terms of HCl was determined from the data to be $n = 0.5$, which is appropriate for the chlorine deposition process involved (see adsorption isotherm [24]). However, this value (i.e., $n = 0.5$) was not speculated prior to determination by fitting the data. The Arrhenius law was used to describe the dependence of the rate constant on temperature:

$$k_{Hg,HCl} = Ae^{-E_a/RT} \quad ((\text{cc/mol})^{1/2}\cdot\text{s}) \qquad (4)$$

The HCl-concentration order of $n = 0.5$ was found to work for all three carbon types. The pre-exponential factor, A $(1/(\text{cc/mol})^{1/2}\cdot\text{s})$, and the activation energy, E_a (J/mol), are presented in Table 2, for each of the three carbon types. Notice the negative activation energy, which is necessary because the mercury-adsorption rate increases with decreasing temperature.

Table 2. Activation energies and rate constants for the three major carbon types.

Parameter	Activated Carbon	Carbon Black	Unburned Carbon
A (1/(cc/mol)$^{1/2}$·s)	1.81 x 10^{-9}	4.54 x 10^{-6}	8.86 x 10^{-9}
E_a (J/mol)	-7.27 x 10^{4}	-3.55 x 10^{4}	-6.15 x 10^{4}

The reaction rate equation and constants shown in Eqs. 3 and 4 and Table 2 were used to predict all of the data measured in the CTF with activated carbon and UBC. There was insufficient data on carbon black to make a comparison between predicted and measured values meaningful. As shown in Figs. 9 and 10, the model reproduces the measured data effectively. The one data point in Fig. 10 that appears to be an outlier was taken under the lowest temperature and chlorine concentration condition, as annotated on the graph (see Fig. 10). At the lowest temperatures and chlorine levels, sorption mechanisms in the system other than the process investigated may begin to play a significant role in terms of measured mercury removal.

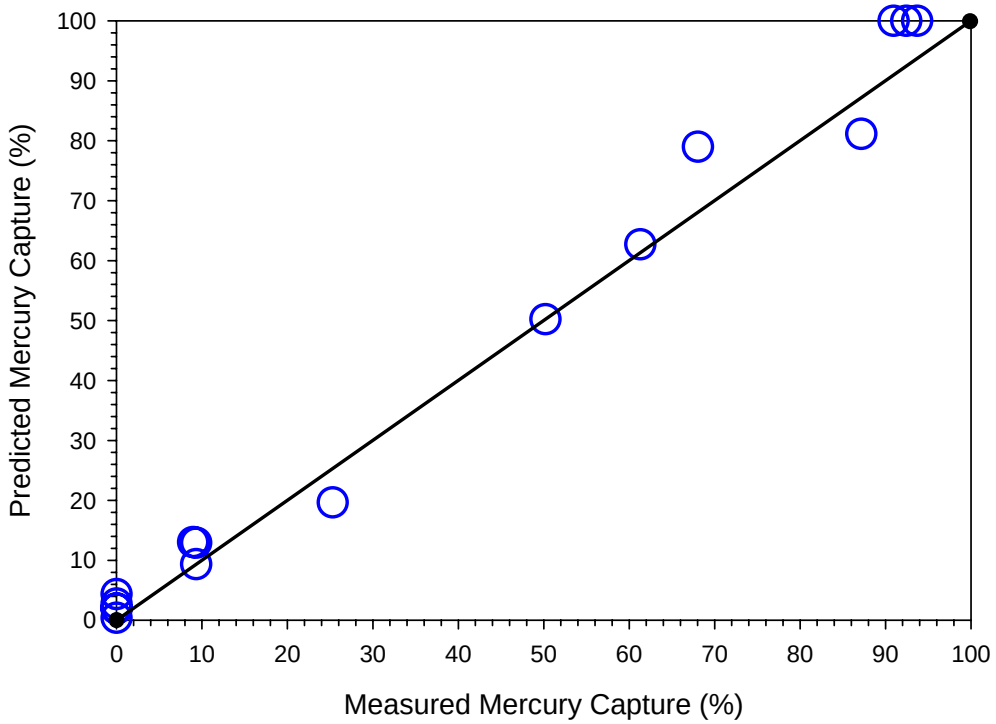


Figure 9. Predicted vs measured mercury capture on activated carbon.

The carbons behaved similarly with respect to mercury adsorption. The major difference between carbon types is the internal surface area of each carbon. Activated carbon, with the highest internal surface area (see Table 1), was most effective at sorbing mercury. The tests in the CTF were designed to investigate adsorption (i.e., the packed-, high-concentration, carbon beds), because observing a combination of oxidation and adsorption mechanisms would have been more difficult to interpret, quantify, and model. Hence, even for the carbons with less surface area, the primary observation was less adsorption. Little oxidation was observed during any of the packed-bed tests. However, in the pilot-scale facility, during tests conducted on the other projects, more carbon surface area resulted in both more adsorption and more oxidation, until the surface area of the carbon was so high that adsorption dominated [3, 22].

The model presented in Eqs. 3 and 4 and rate constants provided in Table 2 describe the rate of adsorption of mercury on three different types of carbon as a function of temperature, time, and flue-gas chlorine content. However, the adsorption rates obtained are also relevant to mercury oxidation, since the mercury-oxidation rate is controlled by carbon catalysis, involving both adsorption of HCl to form chlorinated carbon sites and the desorption of HgCl from the carbon surface. The packed bed environment for these carbon tests did not promote desorption of oxidized mercury back into the flue gas, as do disperse unburned carbon particles contained in a typical coal-derived flue gas. Nevertheless, the rate of elemental mercury adsorption onto the carbon (i.e., chemisorption onto chlorinated carbon sites) should be somewhat similar.

The concentration of carbon and the bed depth in the CTF for these experiments was such that the gas passing through the bed was exposed to much more carbon than would be the case in the disperse flue gas of the pilot plant or even across a baghouse filter cake. This is why adsorption was the main mechanism observed, rather than oxidation, which was a significant advantage in terms of isolating the adsorption mechanism and making quantitative comparisons of different carbon types. Although as much mercury desorption may have occurred in the CTF as in the pilot-scale tests, the extra contact with more carbon made it likely that desorbed oxidized mercury would readsorb.

The CTF setup could have been adjusted and additional experiments performed to produce observations of more oxidation rather than adsorption, by reducing the concentration of carbon (mixing with sand) and the bed depth. However, such experiments would be very difficult to control (these would not be breakthrough tests, which are not representative of the interaction of mercury with carbon in coal-fired power plants), repeatability would be a problem, and uncertainties would be high. In addition, by the time the adsorption mechanism experiments were concluded, there was little motivation to conduct such experiments. At that point, it had been determined that carbon was the only catalyst option identified for duct injection, and activated carbon was by far the most effective at adsorbing mercury, which in hind sight may seem obvious.

Given that activated carbon was already extensively being studied and tested elsewhere for mercury capture via duct injection, and the fact that the mechanisms involving mercury adsorption and oxidation on carbon had been significantly elucidated both in the pilot-scale tests on the other project and in the CTF on this project, it was determined that future efforts and funding on this project should be spent exclusively on the SCR catalyst testing.

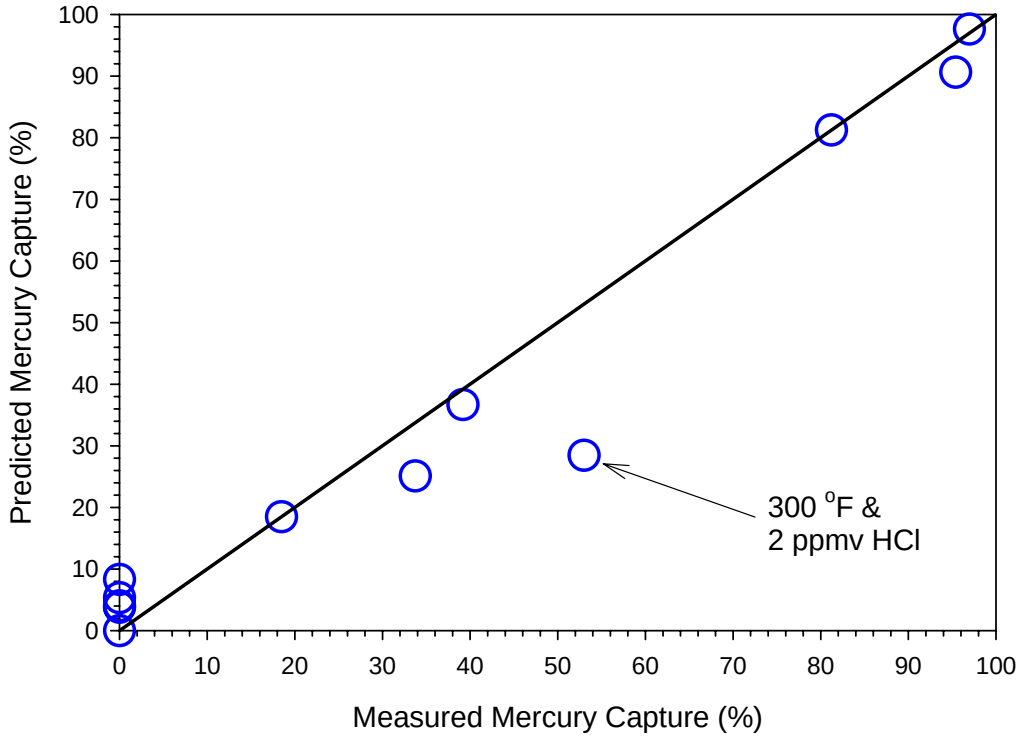


Figure 10. Predicted versus measured mercury capture on unburned carbon.

Examination of Carbon/Calcium Catalytic Synergism

Full- and pilot-scale data have shown that mercury capture is enhanced when both calcium and carbon are present, either in the disperse phase or in a filter cake [3, 6, 14, 15]. As shown in Figs. 11 and 12, mixing calcium with carbon did not appear to enhance the capture of mercury in the CTF at 300 °F. Little mercury desorption took place in the CTF tests, even for the carbon-only experiments conducted in this investigation, which suggests that Ca-enhancement of mercury capture in full-scale units may be associated with the oxidized-mercury desorption step. However, it should be remembered that the sorbents are compared on a common bed-residence time for each sorbent. Hence, the 10% C/ 90% hydrated-lime sorbent, only exposed the simulated flue gas to approximately 10% of the carbon used in the pure activated-carbon tests. Longer-exposure-time (breakthrough) tests might produce a benefit of adding calcium to the bed material, if reactive capture of mercury by calcium occurred. However, the investigation of calcium enhancement of mercury capture funded by DOE on the DOE CBS project [3] at Southern Research has concluded that the dominant impact of calcium on the enhancement of mercury capture in coal-fired boilers is to prevent the release of oxidized mercury back into the gas phase [3, 23]. Several pathways were suggested in a recent publication [23] as possible fundamental mechanisms.

As shown in Fig. 11, mercury adsorption was more effective with activated carbon than with the C/Ca sorbent at 300 °F. With 2 ppmv of HCl, based on only two data points, the C/Ca sorbents appeared to be slightly more effective at higher temperatures, where desorption is more dominant and adsorption less dominant as temperature increases. In general however, no significant

advantage was observed for the C/Ca mixtures, which further suggests that the mercury-capture enhancement mechanism involving calcium was not reactive capture by the calcium.

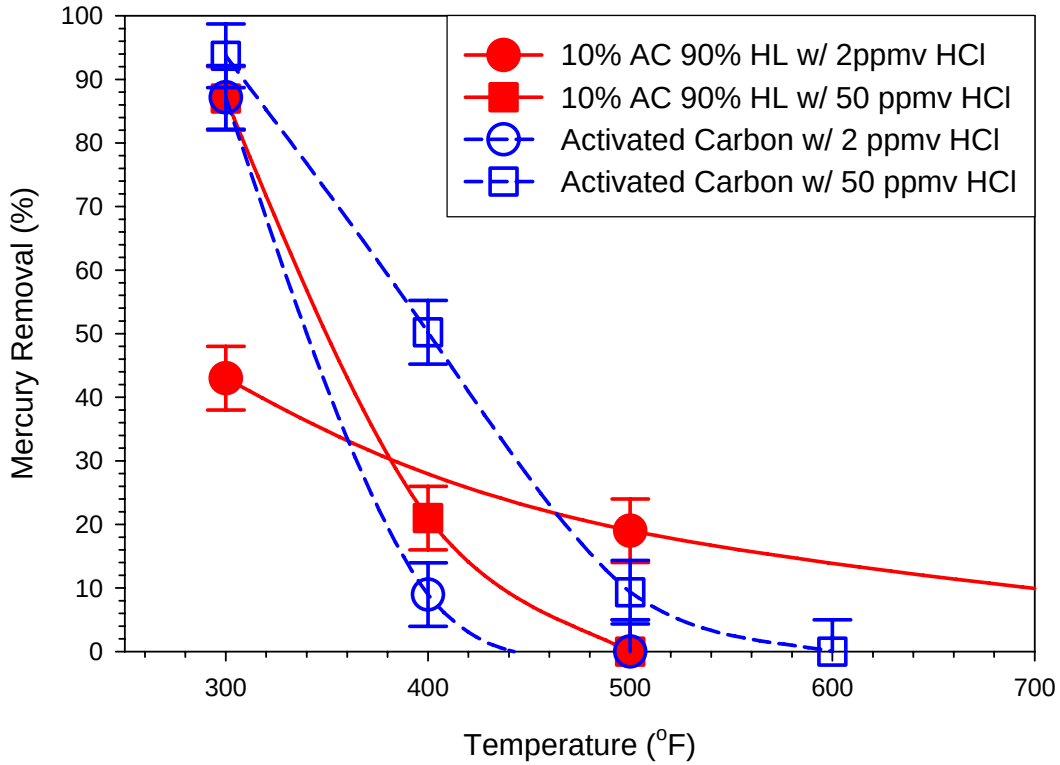


Figure 11. Activated carbon compared with 10% AC/90% HL processed sorbent mixture.

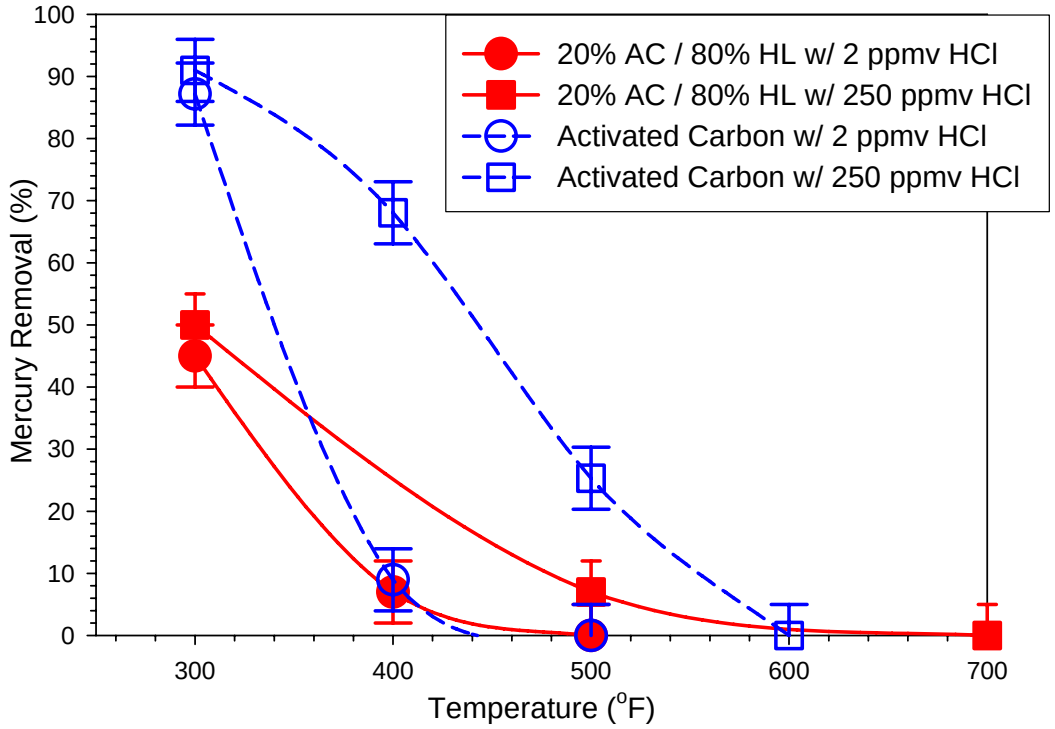


Figure 12. Activated carbon compared with 20% AC/80% HL processed sorbent mixture.

Figure 12 illustrates similar behavior for the 20% C/80% hydrated lime sorbent. Figure 12 illustrates the comparison of mercury capture at 2 and 250 ppmv HCl, whereas Fig. 11 illustrates the comparison of mercury capture at 2 and 50 ppmv HCl. At these high chlorine concentrations, activated carbon appears to be much more effective at sorbing mercury than Ca/C sorbents, even at the higher temperatures. The Ca/C sorbent appears to be much less affected by the presence of such high levels of chlorine. Higher levels of chlorine in the flue gas induce higher levels of chlorine on the carbon surface. Although orders of magnitude lower than the concentration of chlorine in the flue gas, the elevated fraction of chlorinated-carbon sites significantly increases mercury chemisorption. The lower carbon concentration of the calcium-based sorbents limits the number of chlorinated carbon sites that can be formed per unit mass of sorbent. However, the presence of calcium may enhance the chances of oxidized mercury remaining adsorbed once oxidized.

Activated carbon was the most effective carbon mixed with calcium/carbon sorbents, and limestone behaved similarly to hydrated lime, based on findings from the DOE CBS project [3] testing in the CTF, although this was not tested on this project. Based on the findings from the other project, under PRB-like simulated flue gas (i.e., with 2 ppmv HCl) at 300 °F, approximately 86% mercury removal was obtained with a 10% activated carbon/90% limestone mixture, and approximately 60% mercury removal was obtained with a mixture of 10% activated carbon and 90% hydrated lime. However, no repeats were conducted, and so the uncertainty is such that all that could be concluded was that limestone behaved similarly to that of hydrated lime, when mixed with activated carbon. A mercury removal percentage for the same activated carbon/hydrated lime mixture was also reported to remove approximately 15% of the mercury at 400 °F [3]. Finally, the results of a comparison between mixtures of carbon black and hydrated lime with activated carbon and hydrated lime (e.g., 10%/90%) were presented, for the same simulated flue gas. The mixture containing activated carbon removed approximately 41% of the mercury at 300 °F, compared to the sorbent containing carbon black, and the mixture containing activated carbon removed about twice as much (15% compared with 7.5%) as the mixture containing carbon black at 400 °F [3].

SCR Catalyst Investigation

Establishing Basis for Comparing SCR Catalysts

Catalysts for SCRs are designed for NO_x control. However, mercury oxidation is an unintended, beneficial reaction that occurs in the SCR process. Hence, the basis used for the comparison of SCR catalysts in this investigation was equivalent NO_x reduction. The *quantity* of each type of catalyst used was based on the *quantity* needed to obtain a NO_x removal percentage of 70%, at a one-to-one ammonia to NO_x molar ratio, while maintaining an equivalent space velocity for each catalyst type. The temperature, flow, and gas composition to establish the equivalent *quantity* of each catalyst type were slightly different than those used during the mercury oxidation comparison test campaign, the conditions for which resulted in greater than 95% deNO_x, typical of commercial operating units.

A blank quartz reactor was inserted into the gas stream at the beginning of these initial tests to collect baseline data. After sufficient data were collected, the blank reactor was removed and

replaced with the catalyst-packed reactor. An inert nitrogen stream replaced the equivalent ammonia flow required for a one-to-one ammonia-to-NO_x ratio. The catalyst temperature and gas temperature were allowed to reach a steady-state value of 700 °F, and the NO_x concentration was allowed to reach a steady-state value of 300 ppmv. The NO_x value was recorded as the inlet NO_x concentration. Following the inlet NO_x-concentration measurement, the ammonia was turned on. The NO_x concentration was again allowed to reach steady-state, which was taken as the outlet NO_x concentration.

If the $deNO_x$ was greater or less than 70%, the surface area was altered appropriately and retested, until the exact quantity of each catalyst type was obtained that yielded a consistent $deNO_x$ of 70%. Figure 13 illustrates the final validation of equivalent $deNO_x$ (which as shown was performed at conditions yielding ~50% $deNO_x$) for each of the three catalyst samples, one of each catalyst type. This process confirmed that the comparable volume of each catalyst to obtain equal $deNO_x$ resulted in very similar area velocities.

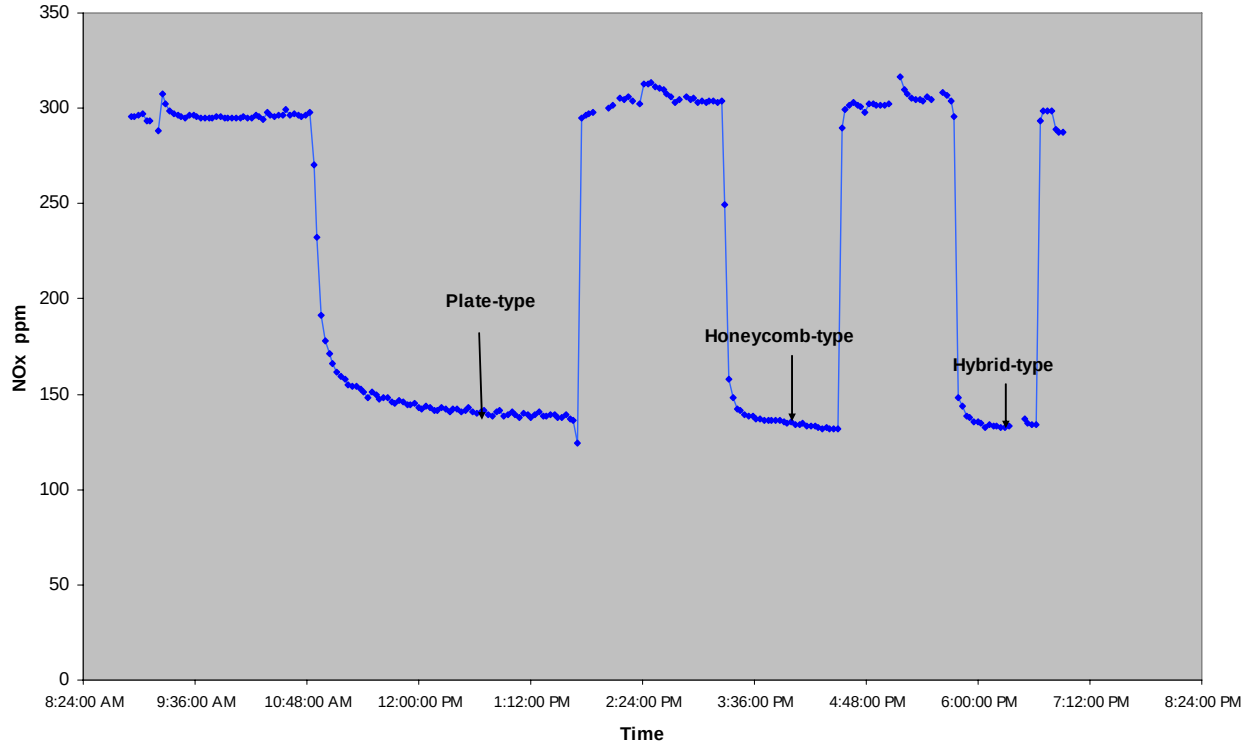


Figure 13. $DeNO_x$-validation establishing equivalent quantities of each catalyst type.

SCR Catalyst Test Method

Initial testing indicated that catalyst samples previously exposed to flue gas for several days, achieved a steady-state adsorption and release of mercury much faster than new catalyst samples. It was also observed that elevating the temperature or HCl concentration caused a rapid desorption of mercury from the catalyst samples, with lesser but continued release of mercury for several days.

The adsorption data from one previously-exposed catalyst sample is presented in Figure 14. The time required for the catalyst to reach steady state was approximately 7 hours (inlet mercury concentration was 10 $\mu g/m^3$).

Table 3 contains the range of parameters tested in this work. Ammonia concentration was maintained at 300 ppmv, which was based on a one-to-one NH_3/NO $deNO_x$ ratio, with 300 ppmv of NO in the flue gas. A number of tests were performed in the absence of ammonia, to examine both the effect of ammonia and to obtain information relative to off-season operation, for those plants that choose to keep their flue gas flowing through the SCR, during the season when $deNO_x$ is not required.

Sulfur trioxide was measured via the controlled-condensation method downstream of the reactor for each test condition, to determine the fraction of SO_2 oxidized across each catalyst type. The area velocity is a good parameter to use to simulate the conditions for use in full-scale units, and so was kept within a range of 4.5 to 8.6 $m^3/hr_{stp,\ wet}/m^2$ for all catalysts and tests performed. This was consistent with the range of SCR catalyst area velocities installed in full-scale units, which typically range from 2.4 to 11.9 $m^3/hr_{stp,\ wet}/m^2$. For reaction systems dominated exclusively by selective catalytic reactions (e.g., SCR reactors), the controlling factor is the contact time with the catalytic surface area, represented by area velocity. Hence, micro-scale SCR tests scale up much better to full-scale systems with area velocity than with space velocity. However, the space velocities in the CTF did ranged from approximately 900 to 2600 hr^{-1}, which is consistent with space velocities for full-scale SCR installations, which are typically >1000 hr^{-1}.

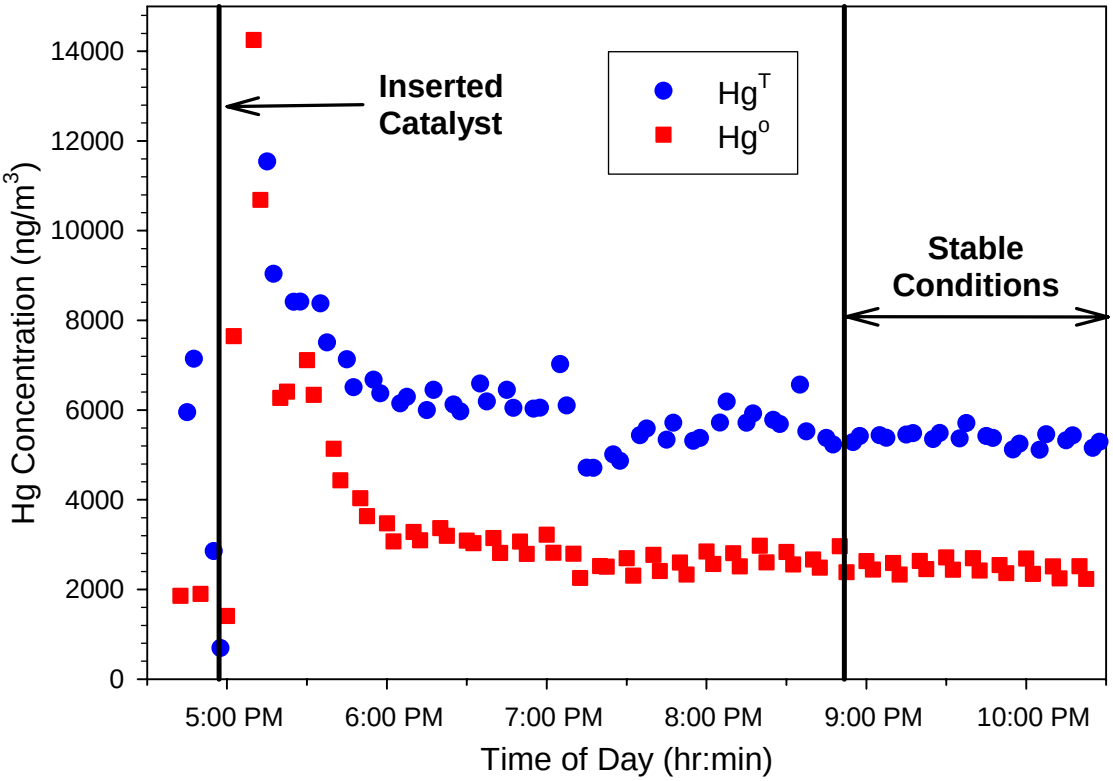


Figure 14. Example of time for previously-exposed catalyst to come to steady state.

Table 3. Test Conditions.

Parameter	Range for Test Campaign
Temperature	650 to 800 °F
Flow Rate	~7.5 slpm
$deNO_x$	>95% $deNO_x$ at $NH_3/NO = 1$
Gas Concentrations (dry basis, other than H_2O)	
N_2	~72% by volume
O_2	~5 % by volume
CO_2	~15% by volume
NO	300 ppmv
NH_3	0.0 ppmv or 300 ppmv
SO_2	500 ppmv or 1000 ppmv
HCl	2 to 100 ppmv
H_2O	~8% by volume
Hg°	5 to 10 $\mu g/m^3$

$DeNO_x$ values were maintained near or above 95% for all of the experiments, where normal $deNO_x$ operation of an SCR was simulated using a one-to-one molar ratio of NH_3 to NO. As shown in Figure 15, these $deNO_x$ values were consistently maintained for each of the three catalyst types tested during the investigation. Figure 15 also illustrates that the HCl concentration had little or no effect on $deNO_x$. The tabular data used to create Figs. 15-20 may be found in Tables A.4 and A.5 in the Appendix.

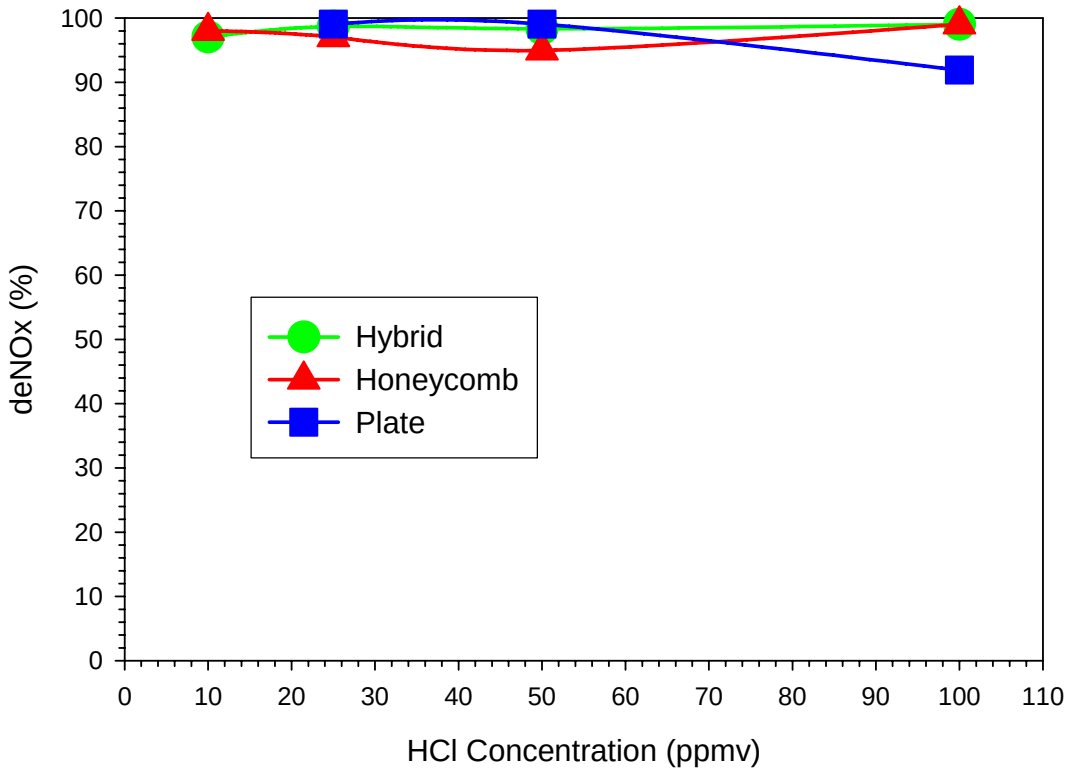


Figure 15. $DeNO_x$ versus HCl concentration conducted at 750 °F.

While the $deNO_x$ for the plate catalyst dips at the 100 ppmv HCl concentration to around 90%, the value for the honeycomb catalyst increases at the HCl concentration in a similar but opposite manner. It is therefore likely that the dip is caused by inconsistency in the test, which for deNOx differences above 90%, can be altered by relatively small changes in the system. Since this was not a $deNO_x$ investigation, the data in Fig. 15 provide sufficient confidence that the $deNO_x$ of each catalyst type reached the required 95% level, for the conditions tested in this work.

Catalyst Comparisons

Initial experiments on flue gas simulated to represent that produced from burning PRB coal (i.e., 500 ppmv SO_2 and 2 ppmv HCl), suggested a significant impact of ammonia on mercury oxidation across SCR catalysts. Tests were performed across the honeycomb catalyst with and without ammonia. Without ammonia (yet with 500 ppmv SO_2), significant mercury oxidation was observed. With ammonia, the measured mercury oxidation across the catalyst was less than 5%. In addition, the effect of SO_2 on mercury oxidation was examined by performing a similar test without SO_2 (but with 300 ppmv NH_3). Without SO_2 present, mercury oxidation was around 5%, not much different than with SO_2. Hence, the presence of SO_2 had little effect on suppressing mercury oxidation, while the presence of ammonia significantly reduced mercury oxidation. As will be shown in subsequent figures and discussion, the impact of ammonia on mercury oxidation was a peculiar function of the low-HCl conditions of the PRB-type flue gas investigated.

Figures 16 through 18 contain graphs illustrating the extent of mercury oxidation across the three different commercial SCR catalysts tested, as a function of HCl concentration in the simulated flue gas. At the inlet of the reactor, (as mentioned in the Experimental section) all of the mercury was in the elemental mercury form, at a concentration of approximately 10 $\mu g/m^3$, which is about the median mercury concentration for the range of coals used in the United States. The vertical axis represents the elemental-to-total vapor-phase mercury ratio at the exit of the reactor. Hence, the top of the graph (i.e., a ratio of 1) represents a condition where no mercury oxidation occurred; while data points near the bottom of each graph represent conditions where significant mercury oxidation occurred.

In addition to the relationship of mercury oxidation with HCl concentration, the figures compare results with and without (wo) NH_3 present for $deNO_x$, at otherwise identical conditions. Unless otherwise noted, all of the data shown in Figs. 16-18 were taken at 700 °F. The standard deviations are provided as error bars, which generally lie within the range of the size of the symbols representing the data points. In cases where error bars are not visible, the standard deviation of the measurement should be assumed to be the width of the data-point symbols themselves.

Figure 16 contains results obtained for the plate catalyst. As expected, the concentration of HCl is a dominating parameter, such that little mercury oxidation is observed with only 2 ppmv of HCl in the flue gas and essentially complete oxidation of the mercury is obtained at 100 ppmv of HCl.

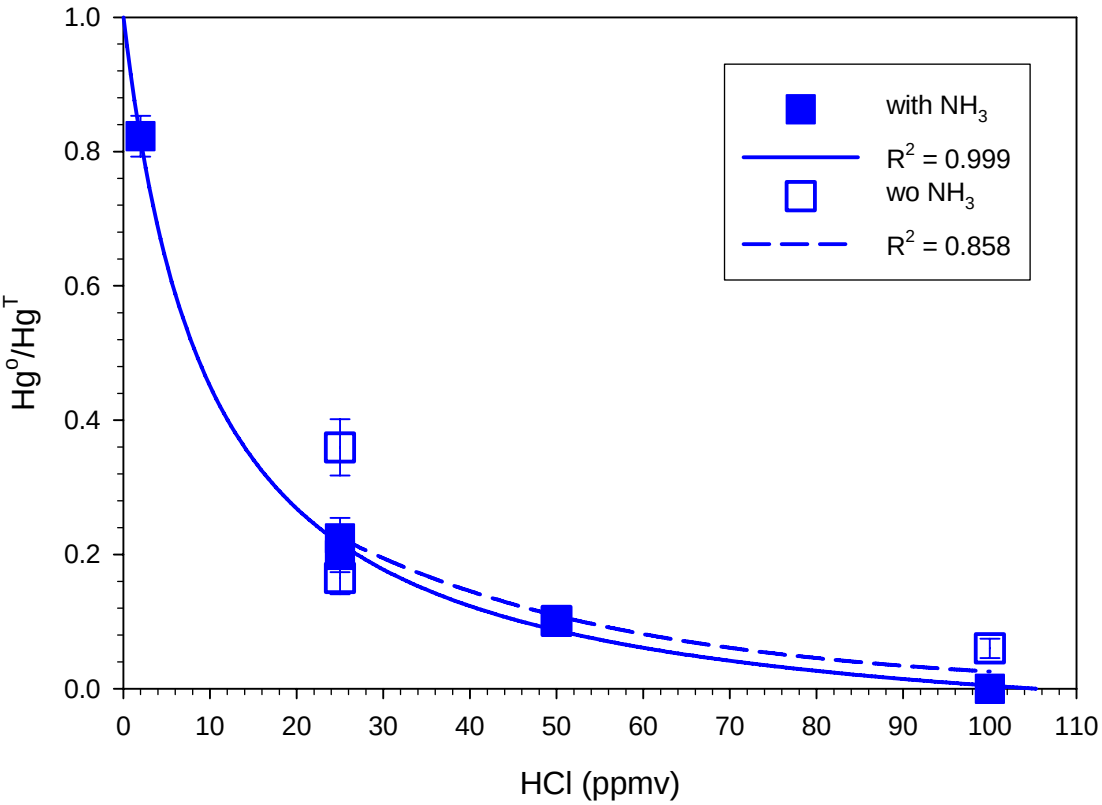


Figure 16. Mercury oxidation across the plate catalyst at 700 °F.

For the range of HCl concentrations tested (i.e., 25 to 100 ppmv HCl), the absence of ammonia appeared to have little effect on mercury oxidation over the plate catalyst. While the results for the hybrid-type SCR catalyst shown in Fig. 17 are very similar to that for the plate catalyst, the absence of ammonia was somewhat of a benefit toward mercury oxidation, in the range of HCl concentrations, 10 to 50 ppmv.

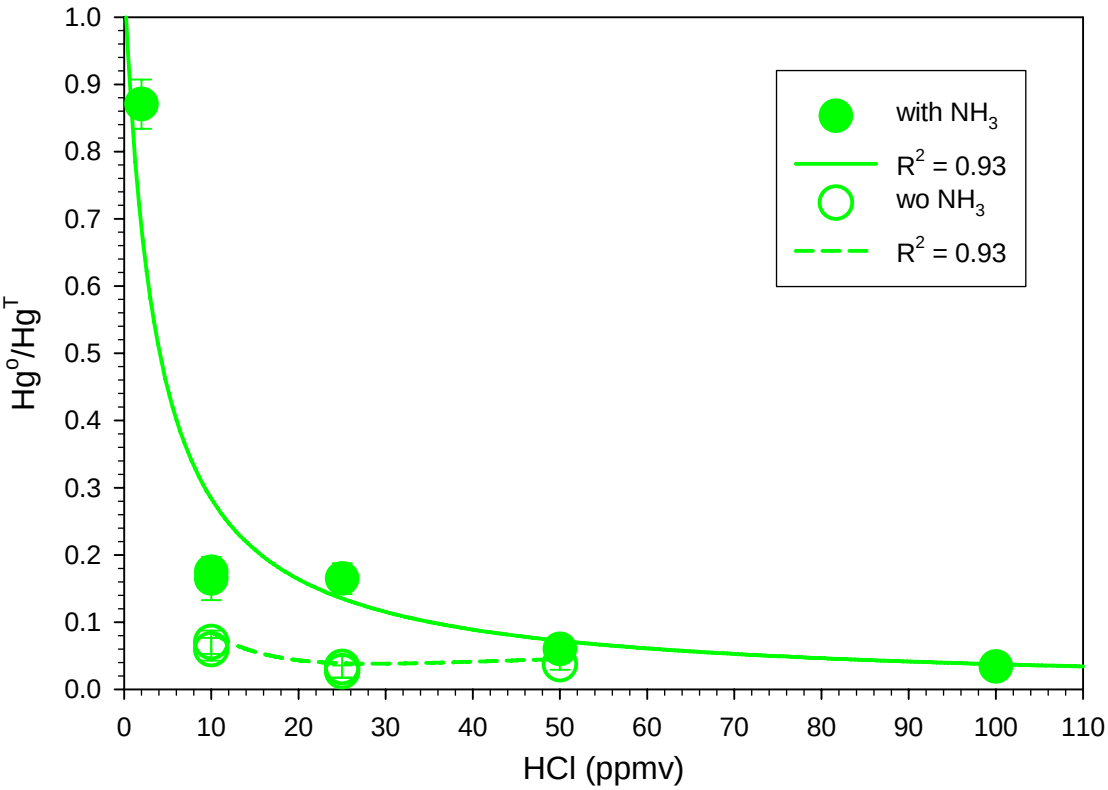


Figure 17. Mercury oxidation across the hybrid catalyst at 700 °F.

As shown in Fig. 18, the honeycomb catalyst performed similar to the plate catalyst, with and without ammonia at 700 °F. However, Fig. 18 also contains two data points obtained while the honeycomb catalyst was maintained at 750 °F, with 2 ppmv HCl in the flue gas. While the two data points appear on the graph to be consistent with what might be expected for data obtained at 700 °F, there is a large difference in mercury oxidation with and without ammonia. It may be that the impact of ammonia on mercury oxidation across SCR catalysts is a phenomenon of importance only at low HCl concentrations.

Within the variation expected between tests, the three catalyst types appear to behave similarly. In fact, the results were similar enough between catalyst types that it is not possible to extract differences between catalyst types from the data obtained in this work.

Mercury oxidation across the plate catalyst with 25 ppmv HCl in the flue gas is compared with and without ammonia in Fig. 19. As shown, the presence of ammonia had little effect on mercury oxidation under these conditions.

Figure 19 also compares the impact of temperature on mercury oxidation across the three commercial catalysts. At 750 °F, all three catalysts produce little if any oxidized mercury in a concentration of 2 ppmv HCl. With 2 ppmv of HCl, both the plate and hybrid catalysts exhibited a significant dependence of mercury oxidation on temperature, yielding significantly more mercury oxidation at 650 °F than at 750 ° F. However, with 25 ppmv of HCl there was little dependence of mercury oxidation on temperature for the plate catalyst, in the range of 650 to 800 °F. Hence, maintaining moderate concentrations of HCl at the SCR reactor inlet may negate

interfering effects of NH_3 and dampen the dependence of mercury oxidation on temperature. The temperature dependence of mercury oxidation shown in Fig. 19 is consistent with thermochemical equilibrium predictions [9]. In other words, it is known [3, 9, 19, 22] that mercury is stable in the elemental form in the furnace of coal-fired boilers and does not start to oxidize until the flue gas cools to approximately 1000 °F or below, but this is dependent on the concentration of oxidant (i.e., HCl) in the flue gas. It is also known [3, 9, 19, 22] that the oxidation of mercury in the flue gas is prevented by an activation energy barrier that prevents the reaction of HCl with mercury from occurring. Catalysts help to overcome this barrier and also to increase the rate of reaction, so that the time necessary for the reaction to occur will be less than the time it takes for the flue gas to pass through the catalytic reactor. However, catalysts do not change the thermodynamic driving forces. Therefore, as has been shown for mercury oxidation across SCR catalysts [9], if the concentration and temperature are such that thermodynamic equilibrium is obtained at a low concentration of reaction product, then little oxidation will occur.

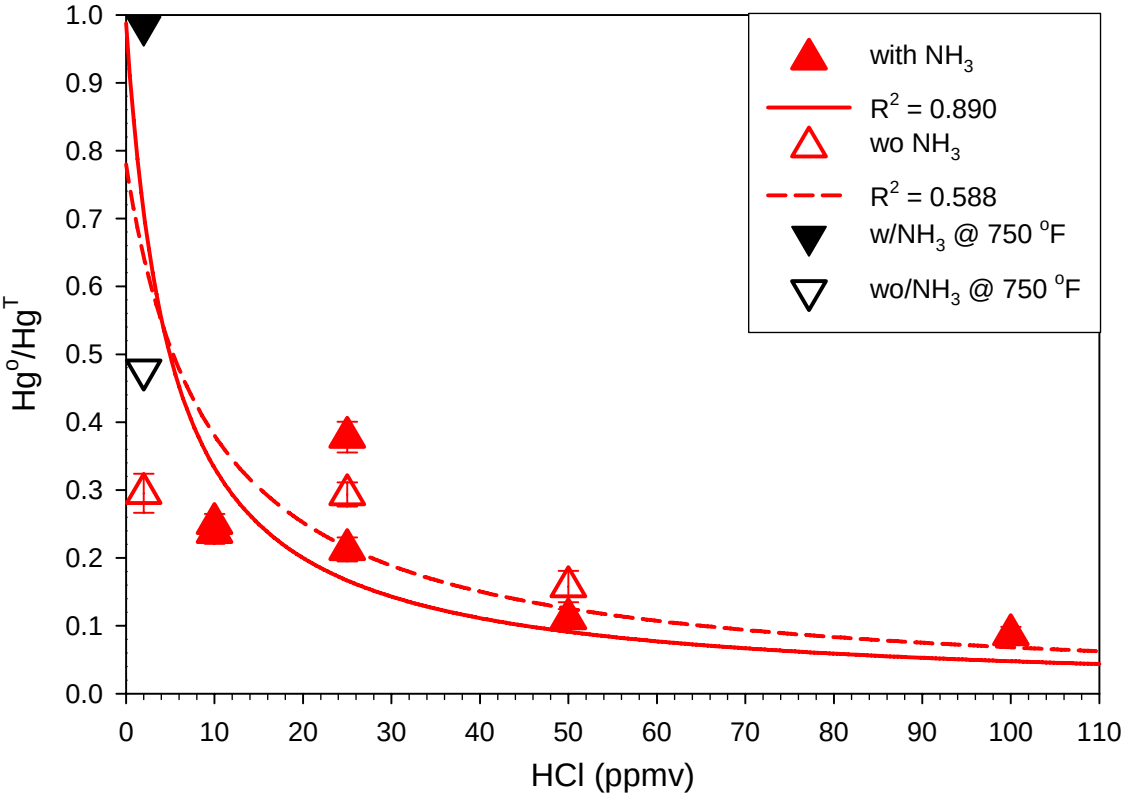


Figure 18. Mercury oxidation across the honeycomb catalyst at 700 °F.

Slipstream [25] and measurements and modeling across full-scale SCRs [11, 26] have also shown this same inhibiting effect of ammonia on mercury oxidation. However, the fluctuations at full-scale power plants (including slipstreams) make it impossible to obtain the detailed quantitative information described in this and other micro-scale investigations.

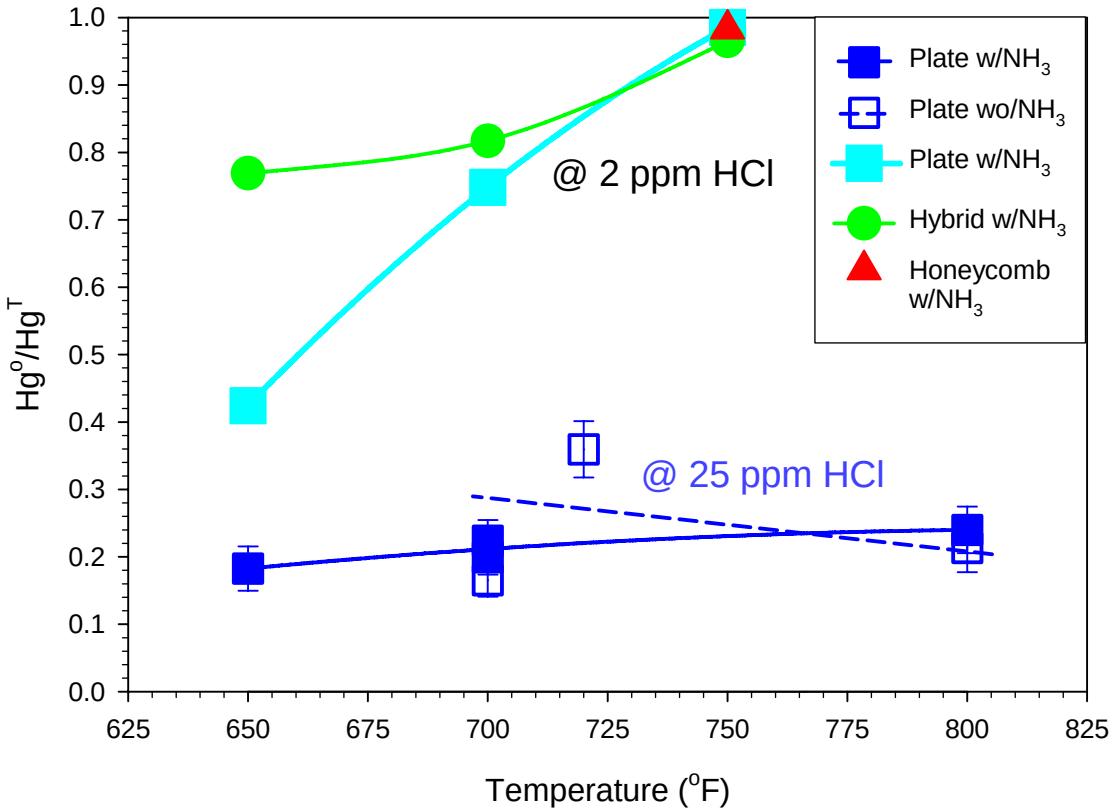


Figure 19. Temperature dependence of mercury oxidation across SCR catalysts.

Figure 20 compares SO_3 formation by each of the three catalysts as a function of HCl concentration. The other conditions were identical for each catalyst, including 700 °F operating temperature and an inlet SO_2 concentration of 1000 ppmv. Concentration differences in HCl did not appear to impact SO_2/SO_3 conversion for any of the three catalyst types. On the other hand, the presence of ammonia appeared to inhibit SO_2/SO_3 conversion. These observations indicate that ammonia competes for adsorption on SCR catalyst sites with SO_2, as it does with mercury. However, it does not appear to compete with HCl for adsorption, which may indicate that HCl is either very weekly adsorbed or that it does not adsorb onto the surface of the catalyst when reacting with mercury, which is adsorbed on the surface of the catalysts.

The data shown in Figure 20 indicate very similar results for the honeycomb and plate catalysts, while the hybrid catalyst behaved somewhat differently. The hybrid catalyst appeared to consist of a more porous substrate, and perhaps that contributed somewhat to the difference in behavior.

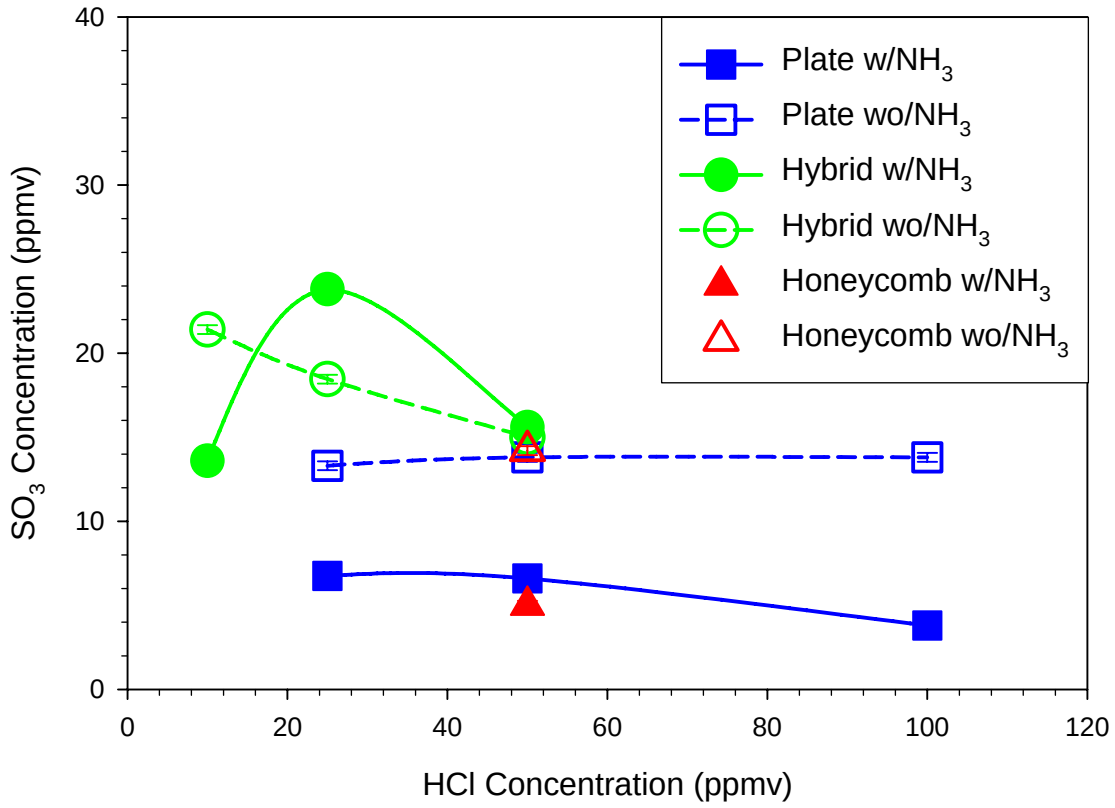


Figure 20. Catalyst comparison for SO_3 formation.

As mentioned above in the Overview, CRF testing that was planned for scaling up and demonstrating catalytic-application methods derived from the bench-scale work were dependent on successful identification of such concepts. As explained earlier, by the time the project was awarded, SCR catalyst testing was added to the project to supplement the developmental work on stationary duct catalysts, which had been developed by DOE with URS. In the course of the project, duct injection of catalysts was narrowed to carbon-based catalysts, which were investigated at bench-scale, helping to elucidate fundamental mechanisms governing their use.

A significant investigation of the catalytic role of carbon (as far as mercury oxidation was concerned) was made in the pilot-scale CRF, which was a partner project to this one, and the findings from this extensive work has been published [23]. However, given the results of the catalyst tests in this work, there was not a good reason to perform any CRF testing. No unique method for using catalysts to mitigate mercury emissions had been identified, and therefore, there was no need to demonstrate it. Consequently, effort for this project was used to investigate the effect of SCR catalysts on mercury oxidation and SO_2 to SO_3 conversion.

Modeling

The SCR data obtained in this work was provided to NEA and REI for their use in modeling. However, by the time the SCR data from this work was finally obtained, the modelers from these organizations had already developed their models, based mostly on full-scale data correlations and their own independent fundamental mechanisms. While they had the data from this work to

compare with their model predictions, at this point, little has been done to incorporate the quantitative information obtained in this work into their models. However, subsequent work at Southern Research Institute, funded by a DOE University Coal Research (UCR) project has used the data obtained in this work, along with additional data obtained in the CTF, to develop an independent fundamental model to predict mercury oxidation and SO_2 to SO_3 conversion across SCR catalysts [12, 13, 27]. In addition to the findings of this work, the UCR project included the effect of CO concentration on mercury oxidation, which was found to inhibit mercury oxidation at low HCl concentrations, typical of PRB coal flue gas [12, 13, 27].

The NEA model [11] based its fundamental mechanism on the adsorption of HCl onto the catalyst surface, along with SO_2 and NH_3, with mercury interacting from the gas phase, while the REI model [26] based its fundamental mechanism on adsorbed mercury, SO_2, and NH_3, with HCl interacting from the gas phase. The new model [12, 13] constructed a fundamental mechanism with all species adsorbed onto the catalyst surface, including both HCl and mercury. In other words, the mercury-oxidation reaction was modeled as taking place between two adsorbed species. However, the HCl was weakly adsorbed. As discussed in this report, it has been definitively shown that mercury strongly adsorbs onto and into SCR catalyst material. The active metal for adsorption is vanadium pentoxide. The model predictions were effective across the range 1 to 50 ppmv HCl, 300 ppmv NO, and up to 900 ppmv SO_2 [12].

CONCLUSIONS

The sorption of mercury by three different carbon types, activated carbon, carbon black, and unburned carbon from coal, have been shown to adsorb mercury according to a first order reaction in mercury, across a temperature range of 300 to 700 °F. Mercury sorption on carbon was found to be half order with respect to the concentration of HCl in the flue gas, which is reflective of the mechanism of adsorption of HCl on carbon sites to form $C{\cdot}Cl^-$ sites (i.e., see sorption isotherms [24]). Rate constants and reaction mechanisms describing the rate of mercury sorption on three carbon types were obtained and validated to be effective at predicting the data obtained in this work.

All three commercial SCR catalysts compared in this investigation, provided by three different manufactures, performed similarly with regard to mercury oxidation and SO_2/SO_3 conversion. In addition, certain general mechanistic observations were made. The presence of ammonia for one-to-one deNO_x may inhibit mercury oxidation at power plants with very low concentrations of HCl in the flue gas, such as might be the case for a plant burning a PRB coal. However, at higher concentrations of HCl in the gas, ammonia had little effect on mercury oxidation across the catalyst.

A larger concern might be that regardless of HCl concentration, the absence of ammonia allowed significantly more SO_2/SO_3 conversion. This is troublesome for two reasons. One, if plants run flue gas through their SCR in the off season to oxidize mercury and capture it in their scrubber, yet turn the ammonia off, because they don't need the deNO_x, then they may significantly increase their SO_3 formation. Secondly, most SCR reactors have multiple layers, the last of which typically do not interact with much ammonia, because most of the deNO_x takes place over the first few layers of catalyst, which means that even with ammonia injection, the lower catalyst

layers will continue to generate SO_3 at a high rate. The presence or absence of SO_2 was found to have little effect on mercury oxidation.

The impact of temperature on mercury oxidation was found to be significant at low HCl concentrations. Mercury oxidation significantly decreased with increasing temperature up to at least 750 °F, with an HCl concentration of only 2 ppmv. However, at higher HCl concentrations, the temperature dependence flattened out. Thermodynamic equilibrium calculations suggest that this phenomenon may be entirely due to the thermodynamic potential for mercury oxide species to form at given temperatures and HCl concentrations [9]. Beyond that, the data obtained in this work clearly indicate that increased mercury oxidation can be obtained across an SCR catalyst with increasing HCl concentrations, regardless of temperature, catalyst type, SO_2/SO_3 concentration, or the presence of ammonia, and without increasing SO_2 conversion.

Lessons Learned:

- When considering duct injection of catalysts or sorbents, laboratory-scale facilities, particularly fixed-bed experiments, operate at conditions that are much less representative of full-scale units than slipstream or pilot-scale units, such as the CRF, which possess a temperature-time profile that matches that of full-scale units.
- On the other hand, when considering testing of SCR catalysts, laboratory-scale facilities, such as the Catalyst Test Facility (CTF) used in this work, provide the most representative (and easy to control and measure) conditions of any test facility.
- The only duct-injection catalyst identified in this work was carbon, as a catalyst/sorbent hybrid.
- The most effective carbon type identified as an adsorbent or catalyst, alone or in a mixture with hydrated lime or limestone, was activated carbon, which is being widely used to remove mercury from existing coal-fired utilities.
- Elemental mercury is adsorbed onto SCR catalysts.
- SO_2 is adsorbed on to SCR catalysts.
- NH3 is adsorbed onto SCR catalysts and competes for vanadium pentoxide active sites with mercury, and SO_2.
- HCl may be adsorbed onto SCR catalysts as well, but mercury, ammonia, and sulfur dioxide adsorption on SCR catalysts is much stronger.
- The presence of ammonia (i.e., in a normal 1-to-1 molar ratio of NO to NH_3) inhibits the oxidation of mercury across SCR catalysts, at low-HCl concentrations, consistent with low-chlorine PRB coals.
- At higher concentrations of HCl, ammonia has little effect on mercury oxidation.
- Ammonia was found to inhibit SO_2-to-SO_3 conversion across SCR catalysts, regardless of the HCl concentration.
- Concentration differences in HCl appeared to have little effect on SO_2-to-SO_3 conversion, at HCl concentrations between 10 ppmv and 100 ppmv.
- At low, PRB-levels of HCl, mercury oxidation increased with increasing temperature, from 650 °F to 800 °F. At higher concentrations of HCl, temperature differences within this range of temperatures had little effect on mercury oxidation.
- In addition to the information described in this list, because of the development that took place on this project in the ability to conduct mercury oxidation and SO_2-to-SO_3 conversion

tests across SCR catalysts, much more data and more precise and meaningful results were obtained during subsequent investigations in this area, particularly as part of the DOE UCR project [23]. As a result of this subsequent work, combined with the data obtained in this work, a model describing mercury and SO_2 oxidation across SCR catalysts was developed, based on strongly adsorbed mercury, ammonia, and sulfur dioxide, and weakly adsorbed HCl, which model has been shown to effectively predict our data [23].

ACKNOWLEDGEMENTS

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APPENDIX – DATA

Table A.1. Mercury Removal for different carbon types, with 2 and 50 ppmv HCl.

Material Type	HCl Concentration (ppmv)	Temperature (°F)	Mercury Removal (%)
Activated Carbon	2	300	87.2
Activated Carbon	2	400	9.0
Activated Carbon	2	500	0.0
Carbon Black	2	300	53.1
Carbon Black	2	400	0.0
Unburned Carbon	2	300	53.0
Unburned Carbon	2	400	0.0
Activated Carbon	50	300	93.7
Activated Carbon	50	400	50.2
Activated Carbon	50	500	9.3
Activated Carbon	50	600	0.0
Unburned Carbon	50	300	81.2
Unburned Carbon	50	400	18.5
Unburned Carbon	50	500	0.0
Carbon Black	50	300	40.8
Carbon Black	50	400	14.1
Carbon Black	50	500	0.0

Table A.2. Mercury Removal for different carbon types, with 100 and 250 ppmv HCl.

Material Type	HCl Concentration (ppmv)	Temperature (°F)	Mercury Removal (%)
Activated Carbon	100	300	92.4
Activated Carbon	100	400	61.3
Activated Carbon	100	500	9.3
Activated Carbon	100	600	0.0
Unburned Carbon	100	300	95.4
Unburned Carbon	100	400	33.8
Unburned Carbon	100	500	0.0
Carbon Black	100	300	32.6
Carbon Black	100	400	1.7
Carbon Black	100	500	0.0
Acetylene Soot	100	300	37.2
Activated Carbon	250	300	91.0
Activated Carbon	250	400	68.0
Activated Carbon	250	500	25.3
Activated Carbon	250	600	0.0
Unburned Carbon	250	300	97.0
Unburned Carbon	250	400	39.2
Unburned Carbon	250	500	0.0
Carbon Black	250	300	78.4
Carbon Black	250	400	1.1
Carbon Black	250	500	0.0

Table A.3. Mercury Removal for different carbon/lime mixtures.

Material Type*	HCl Concentration (ppmv)	Temperature (°F)	Mercury Removal (%)
10% C / 90% HL	2	300	43.0
10% C / 90% HL	2	500	19.0
10% C / 90% HL	2	900	3.0
10% C / 90% HL	50	300	87.0
10% C / 90% HL	50	400	21.0
10% C / 90% HL	50	500	0.0
20% C / 80% HL	2	300	45.0
20% C / 80% HL	2	400	7.0
20% C / 80% HL	2	500	0.0
20% C / 80% HL	250	300	50.0
20% C / 80% HL	250	500	7.0
20% C / 80% HL	250	700	0.0

* C = Activated Carbon, HL = Hydrated Lime

Table A.4. $DeNO_x$ data for SCR Catalysts.

Catalyst Type	Temperature (°F)	HCl Concentration (ppmv)	NH_3 Concentration (ppmv)	$DeNO_x$ (%)
Plate	750	25	300	99.0
Plate	750	50	300	99.0
Plate	750	100	300	91.9
Honeycomb	750	10	300	98.0
Honeycomb	750	25	300	97.0
Honeycomb	750	50	300	94.9
Honeycomb	750	100	300	99.0
Hybrid	750	10	300	97.0
Hybrid	750	25	300	98.7
Hybrid	750	50	300	98.3
Hybrid	750	100	300	99.0

Table A.5. Data of Hg- and SO_2-oxidation across SCR Catalysts.

Catalyst Type	Temperature (°F)	HCl Concentration (ppmv)	Ammonia Concentration (ppmv)	Hg^0/Hg^T	SO_3 Generated (ppmv)
Plate	700	2	300	0.823 +/- 0.031	---
Plate	700	25	300	0.199 +/- 0.026	---
Plate	700	25	300	0.224 +/- 0.031	6.8 +/- 0.3
Plate	700	50	300	0.100 +/- 0.018	6.6 +/- 0.3
Plate	700	100	300	0.000 +/- 0.000	3.8 +/- 0.3
Plate	700	25	0.0	0.165 +/- 0.025	13.3 +/- 0.3
Plate	720	25	0.0	0.359 +/- 0.042	---
Plate	700	50	0.0	0.101 +/- 0.012	13.8 +/- 0.3
Plate	700	100	0.0	0.060 +/- 0.014	13.8 +/- 0.2
Hybrid	700	2	300	0.871 +/- 0.037	---
Hybrid	700	10	300	0.175 +/- 0.019	---
Hybrid	700	10	300	0.165 +/- 0.032	13.6 +/- 0.3
Hybrid	700	25	300	0.165 +/- 0.032	23.8 +/- 0.3
Hybrid	700	50	300	0.060 +/- 0.010	15.6 +/- 0.3
Hybrid	700	100	300	0.034 +/- 0.010	---
Hybrid	700	10	0.0	0.070 +/- 0.017	---
Hybrid	700	10	0.0	0.060 +/- 0.017	21.4 +/- 0.3
Hybrid	700	25	0.0	0.033 +/- 0.016	---
Hybrid	700	25	0.0	0.027 +/- 0.009	18.5 +/- 0.3
Hybrid	700	50	0.0	0.037 +/- 0.008	15.0 +/- 0.3

Table A.5 – Continued. Data of Hg- and SO_2-oxidation across SCR Catalysts.

Catalyst Type	Temperature (°F)	HCl Concentration (ppmv)	Ammonia Concentration (ppmv)	Hg^0/Hg^T	SO_3 Generated (ppmv)
Honeycomb	700	10	300	0.251 +/- 0.014	---
Honeycomb	700	10	300	0.238 +/- 0.017	---
Honeycomb	700	25	300	0.212 +/- 0.018	---
Honeycomb	700	25	300	0.378 +/- 0.023	---
Honeycomb	700	50	300	0.111 +/- 0.014	5.0 +/- 0.2
Honeycomb	700	100	300	0.087 +/- 0.011	---
Honeycomb	750	2	300	0.983	---
Honeycomb	700	2	0.0	0.295 +/- 0.029	---
Honeycomb	700	25	0.0	0.293 +/- 0.018	---
Honeycomb	700	50	0.0	0.158 +/-0.023	14.2 +/- 0.2
Honeycomb	750	2	0.0	0.476	---
Plate	650	25	300	0.183 +/- 0.033	---
Plate	800	25	300	0.240 +/- 0.035	---
Plate	800	25	0.0	0.213 +/- 0.036	---
Plate	700	2	300	0.748 +/- 0.026	---
Plate	750	2	300	0.985 +/- 0.015	---
Plate	650	2	300	0.424 +/- 0.022	---
Hybrid	650	2	300	0.769 +/- 0.020	---
Hybrid	750	2	300	0.965 +/- 0.020	---